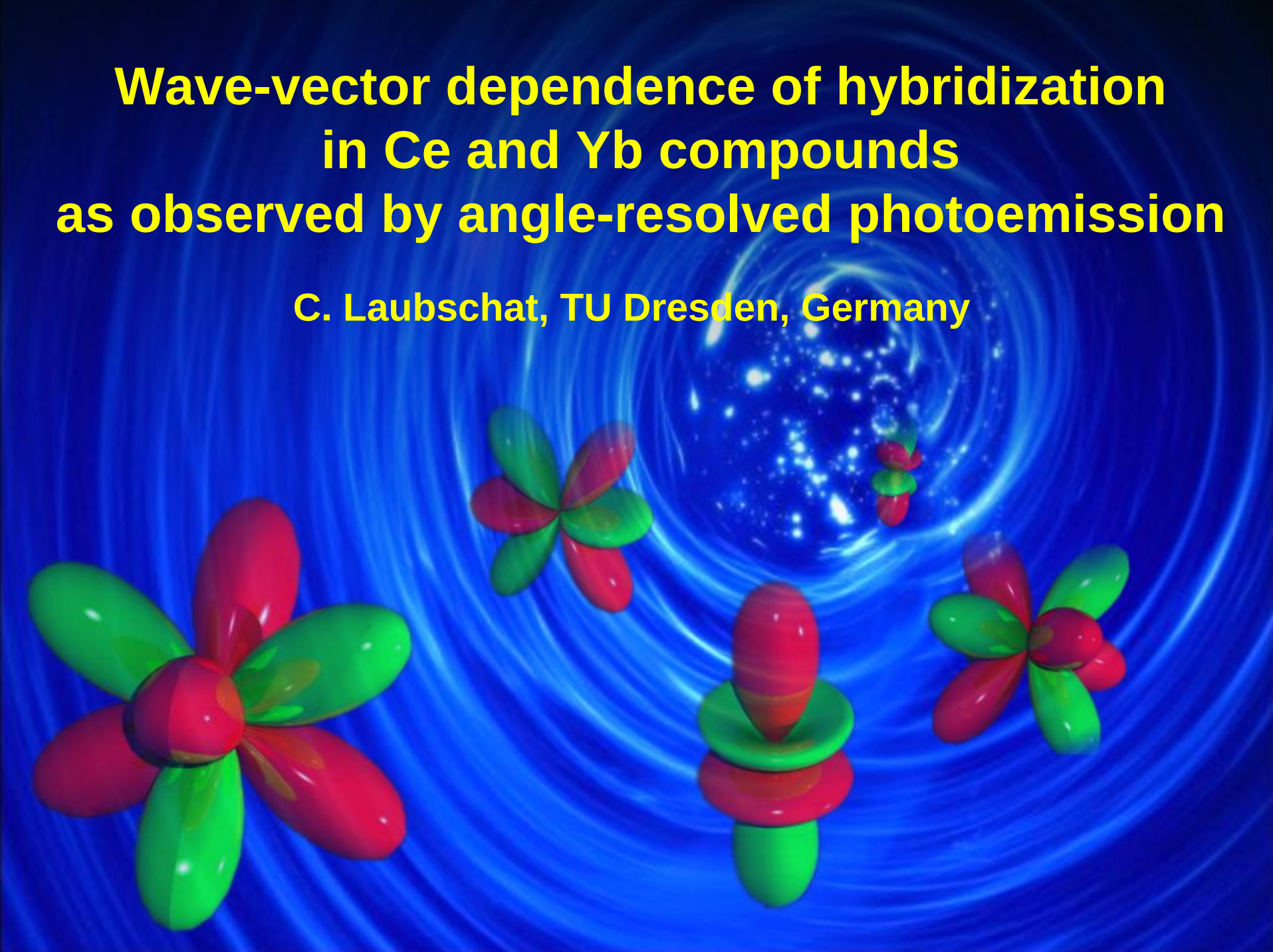
Wave-vector dependence of hybridization in Ce and Yb compounds as observed by angle-resolved photoemission

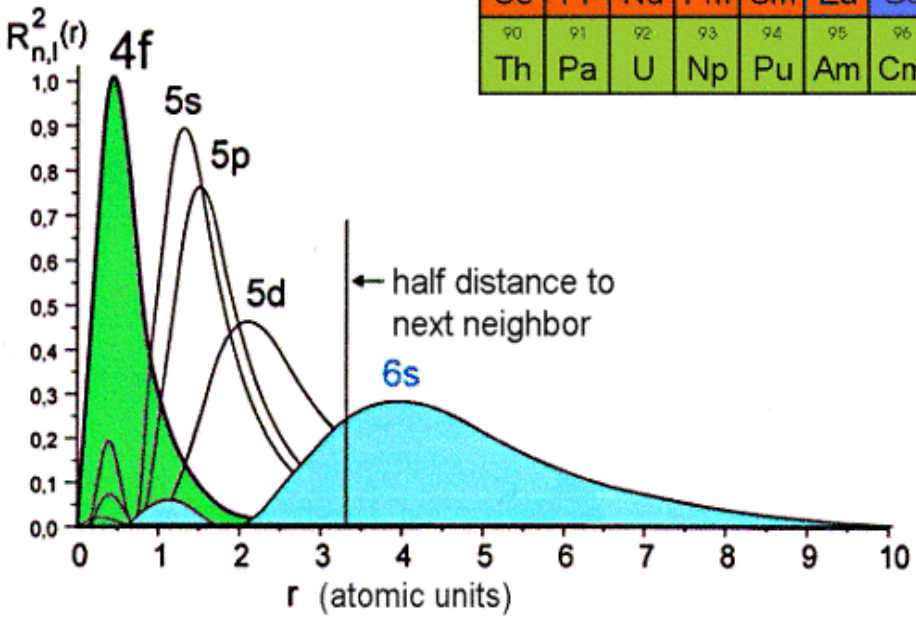
C. Laubschat, TU Dresden, Germany



Periodic System of the Elements

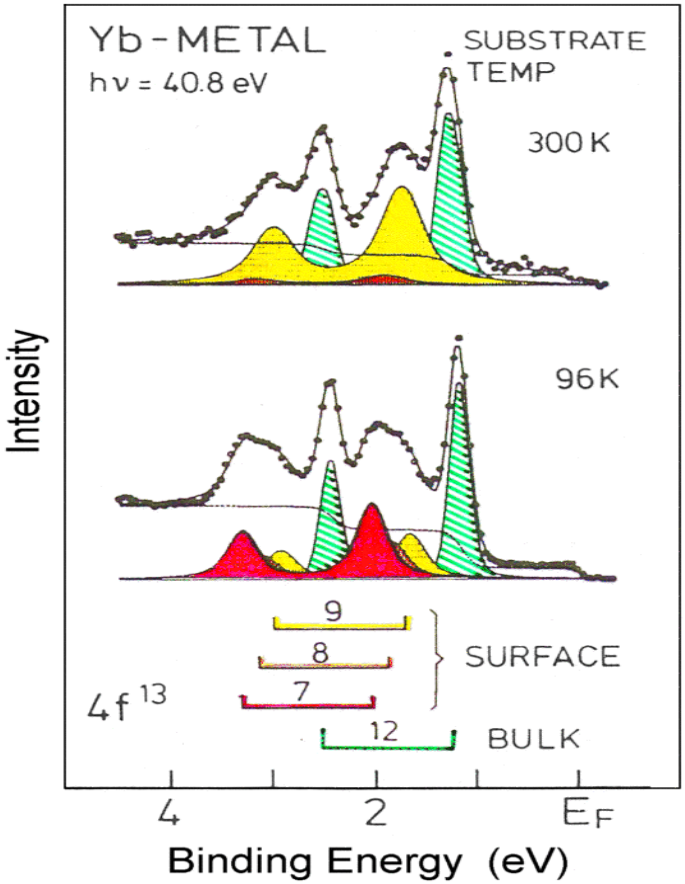
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Ti	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

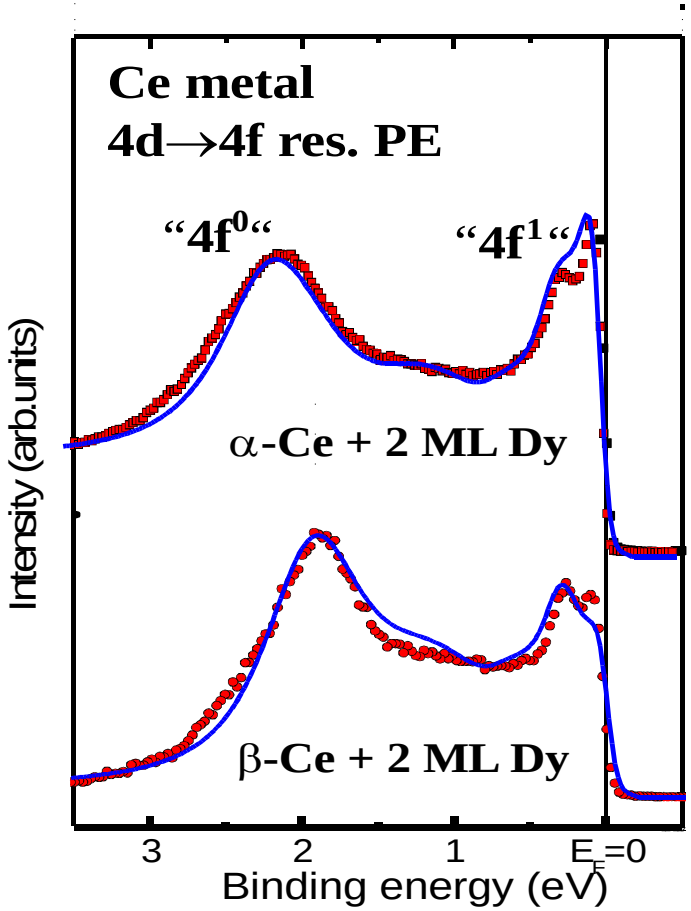


rare-earth elements:

- filling of 4f states
- no f-f overlap
- localized behavior in solid state
- hopping interaction with valence electrons may lead to heavy-fermion behavior



- non-interacting f-states: energy position of atomic-like $4f^{n-1}$ final-state multiplets described within thermo-chemical models



- interacting f-states: coexistence of different final-states described within the Single Impurity Anderson Model (SIAM)

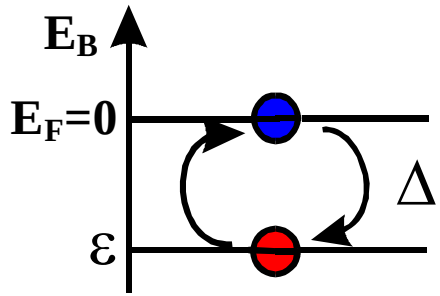
The (Periodic) Anderson Model (PAM) :

$$H = \sum_{\mathbf{k},\sigma} \varepsilon(\mathbf{k}) d^{+}_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma} + \sum_{\mathbf{k},\sigma} \varepsilon_f(\mathbf{k}) f^{+}_{\mathbf{k}\sigma} f_{\mathbf{k}\sigma}$$

$$+ \frac{U_{ff}}{2} \sum_{i,\sigma} n^{f}_{i,\sigma} n^{f}_{i,-\sigma}$$

$$+ \sum_{\mathbf{k},\sigma} V_{\mathbf{k}}(\varepsilon) \left(d^{+}_{\mathbf{k}\sigma} f_{\mathbf{k}\sigma} + f^{+}_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma} \right)$$

Imer*-approach to Single-Impurity Anderson Model



Simplest case: f-state (ε) interacts (Δ) with only one vb-state at E_F (Imer)

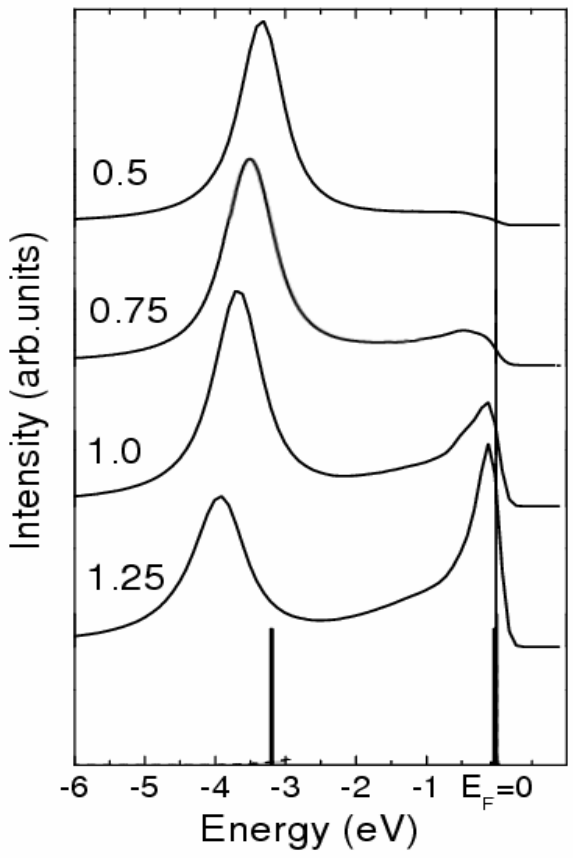
$$|4f^0\rangle := \begin{pmatrix}1\\0\end{pmatrix}, \quad |4f^1\rangle := \begin{pmatrix}0\\1\end{pmatrix}, \quad H := \begin{bmatrix}0 & \Delta\\ \Delta & \varepsilon\end{bmatrix}$$

Diagonalization:

$$|e\rangle := \begin{pmatrix}e_0\\e_1\end{pmatrix}, \quad |g\rangle := \begin{pmatrix}g_0\\g_1\end{pmatrix}, \quad H' := \begin{bmatrix}E_e & 0\\ 0 & E_g\end{bmatrix}$$

$$E_e - E_g = \sqrt{\varepsilon^2 + 4\Delta^2}$$

$$I_g/I_e \cong \frac{\Delta^2}{\varepsilon^2 - \Delta^2} \qquad n_f = 1 - \frac{\Delta^2}{\varepsilon^2}$$



- J.-M. Imer & E. Wouilloud, Z. Phys. B 66, 133 (1987)

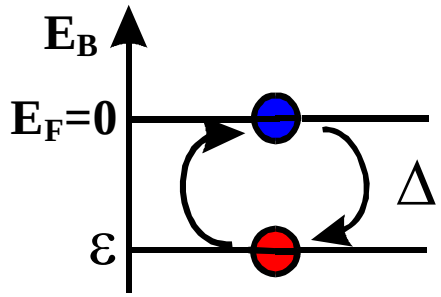
Improved-approach*:

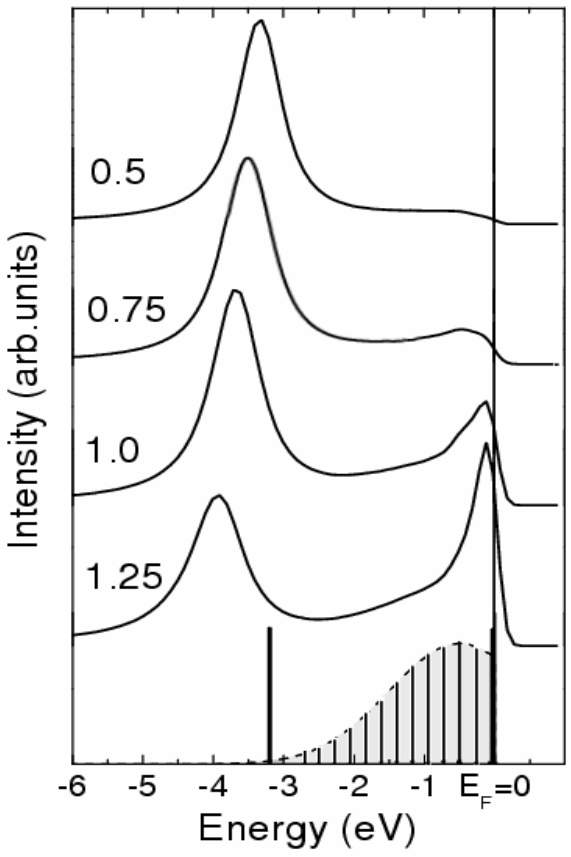
Consideration of :

- density of states modelled by discrete valence band states
- spin-orbit interaction
- double occupation of f-state

results in

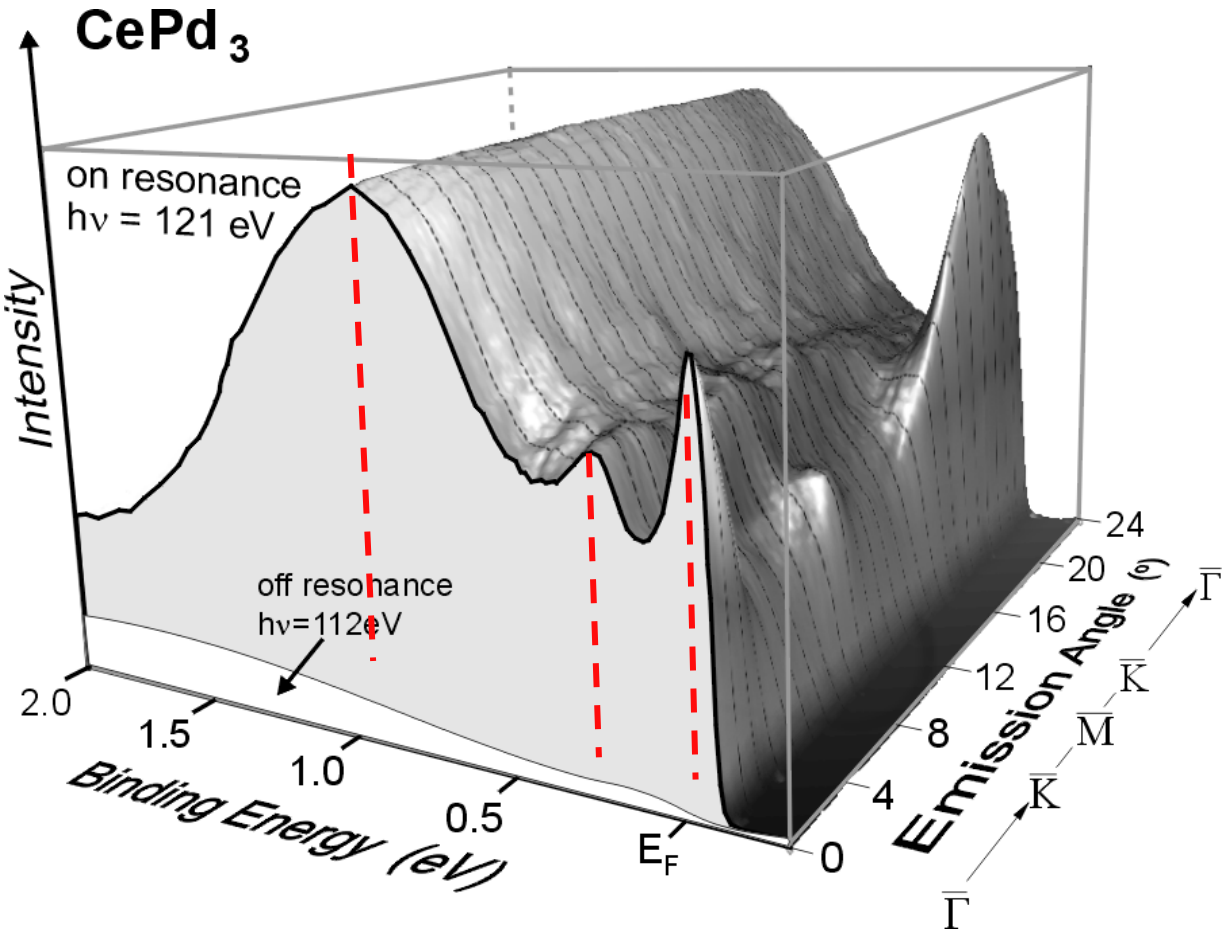
- almost perfect agreement with descriptions in the light of the Schoenhammer-Gunnarsson approach





*Hayn, Kucherenko *et al.*, PRB 64, 115106 (2001)

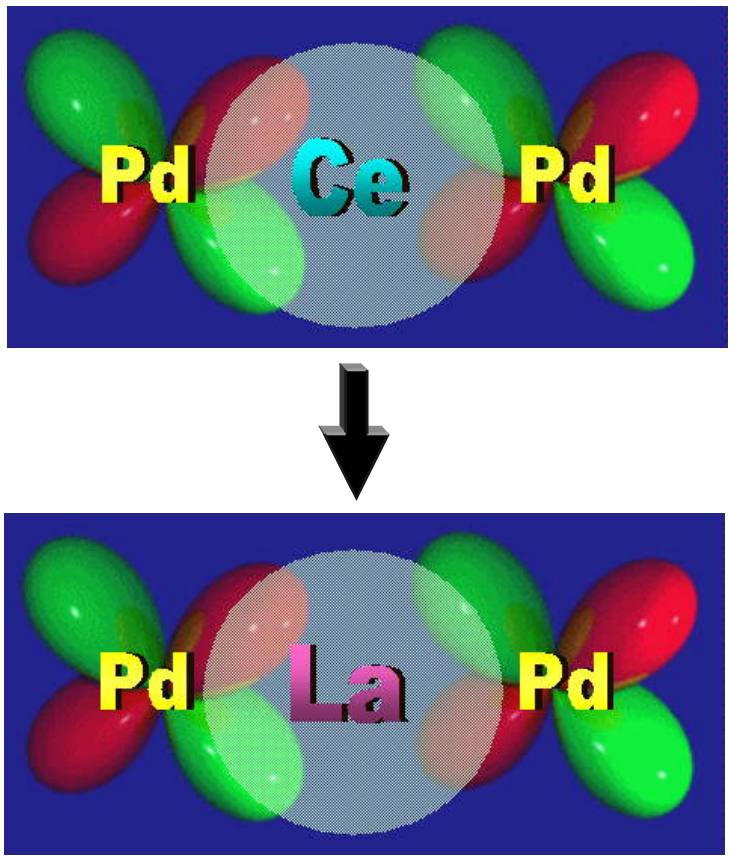
Angle-resolved resonant photoemission: $CePd_3(111)$

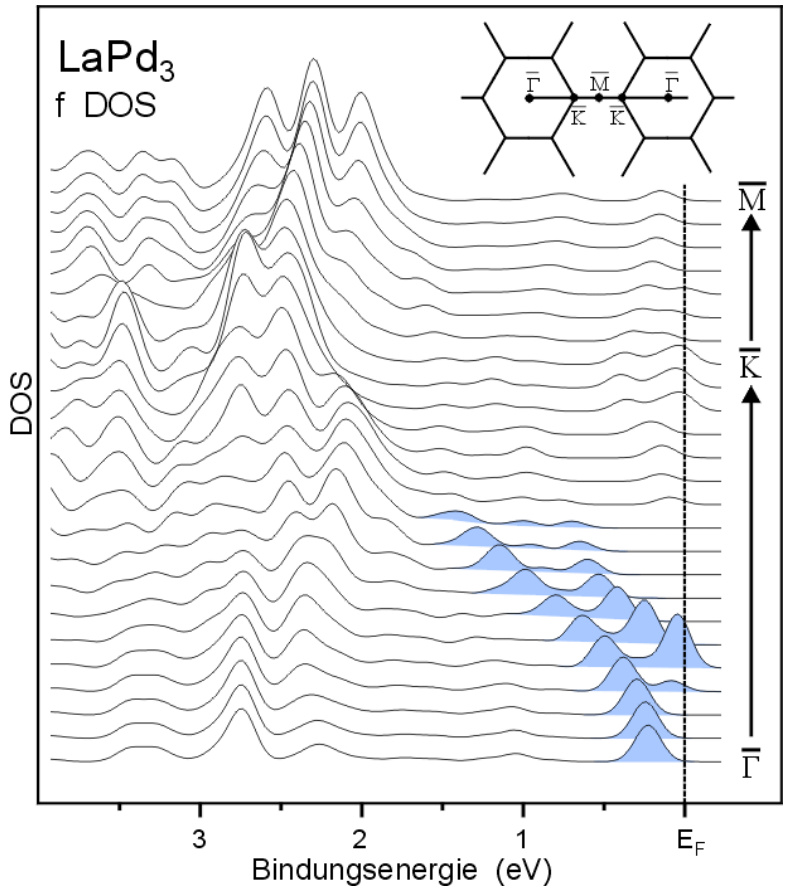


Wave-vector dependent intensity variations of Fermi-level peak!

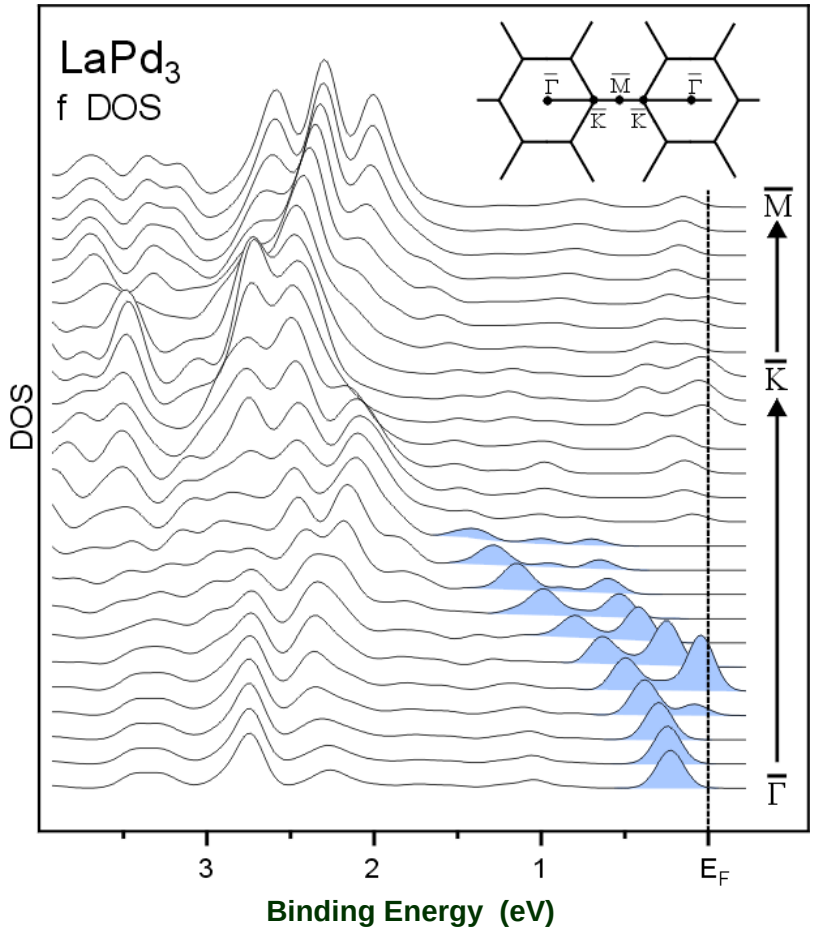
Improved approach:

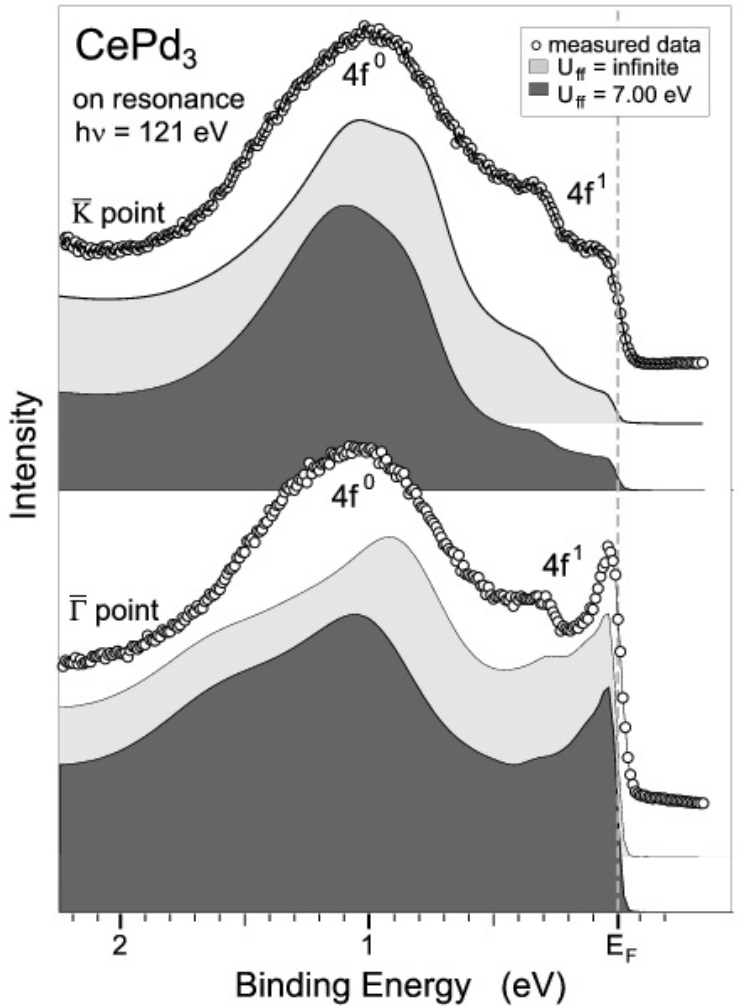
- Assumption of k-conservation upon hybridization
- Application of the model to a *k*-resolved partial density of states !





$CePd_3$: Analysis within PAM

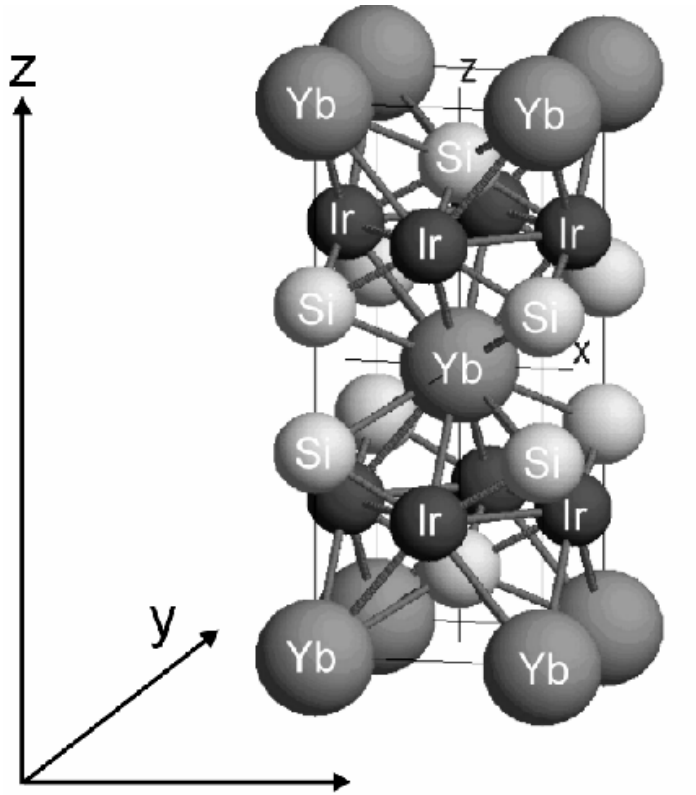




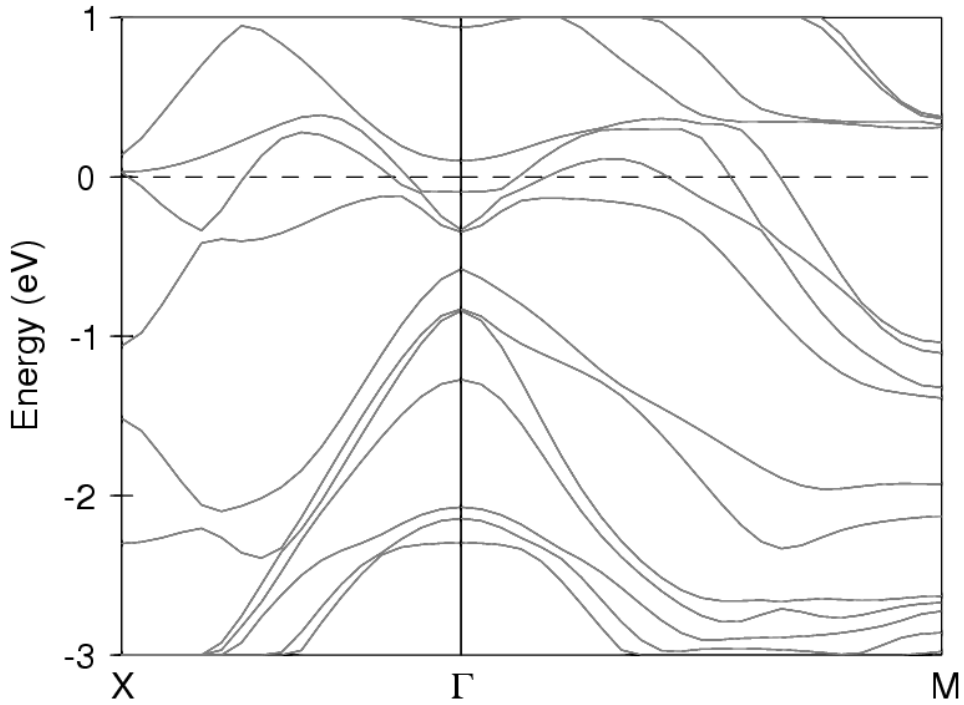
f-character of Pd-derived valence bands at the La (Ce) site

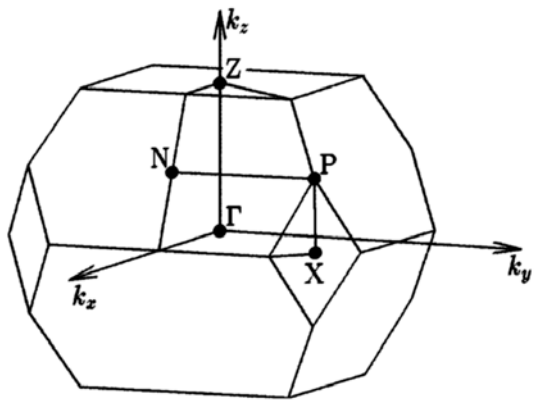
Fermi level crossings of bands lead to strong hybridization at the respective k-points

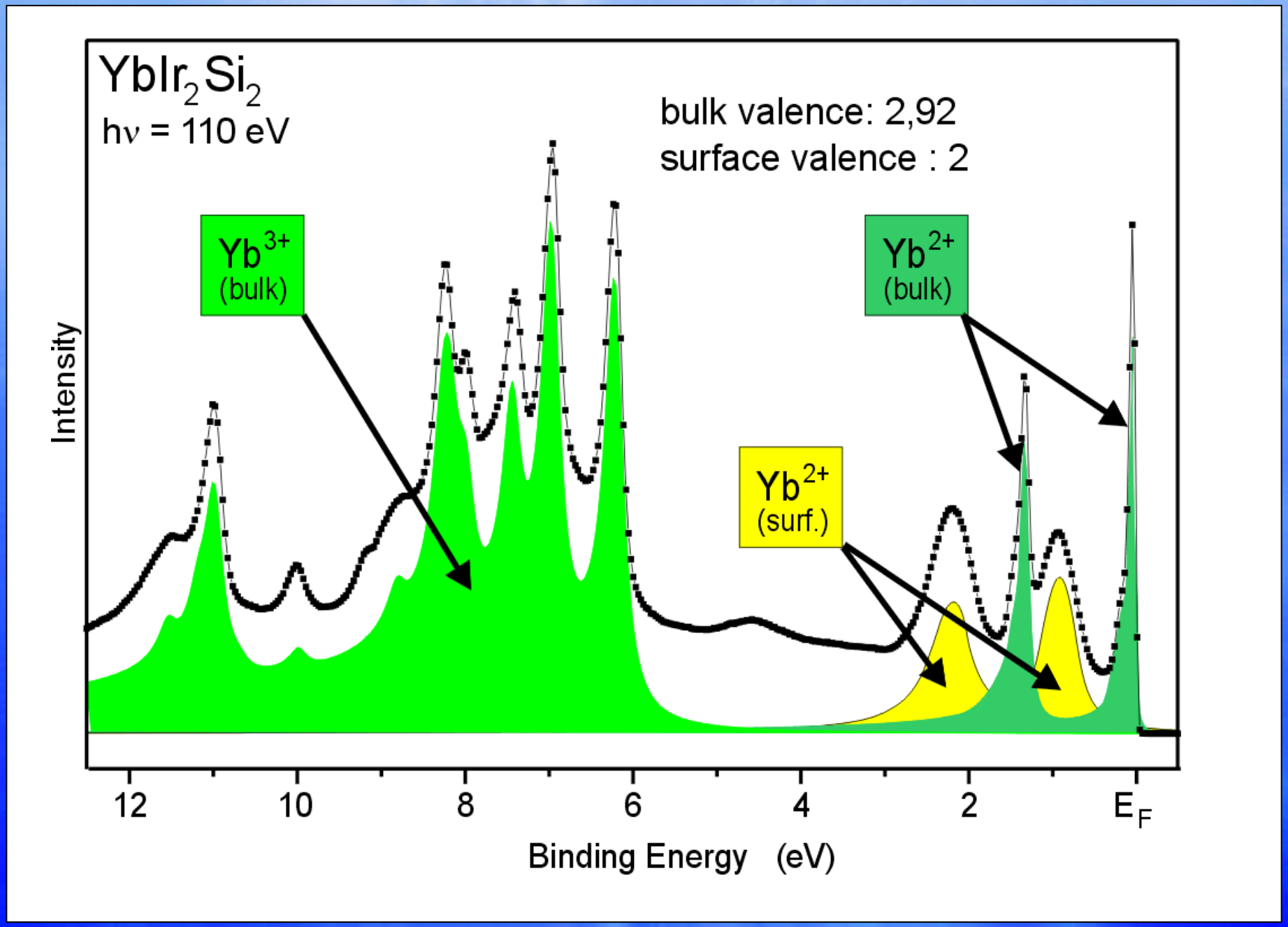
Heavy-Fermion system $YbIr_2Si_2$



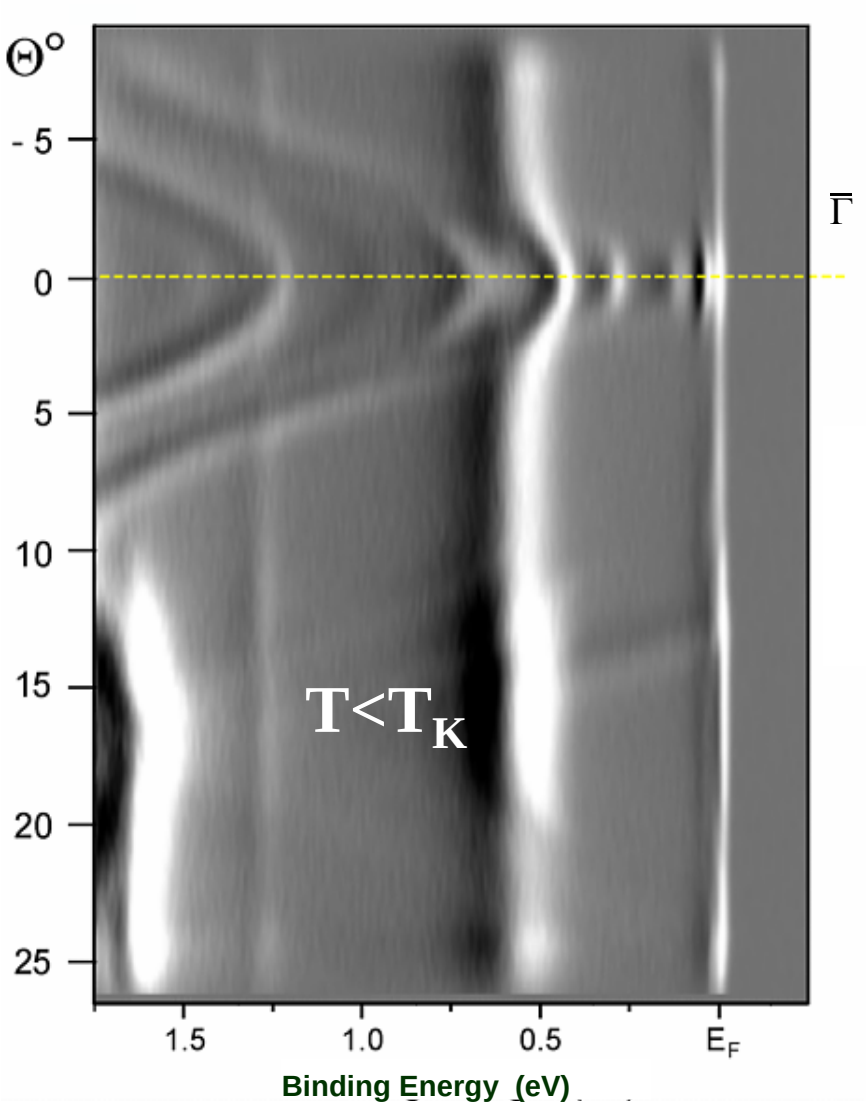
a = 403.5 (1) pm
c = 983.0 (3) pm
z (Si)= 0.3788 (6)
Space group I4/mmm
(Nr. 139) ThCr2Si2 Type

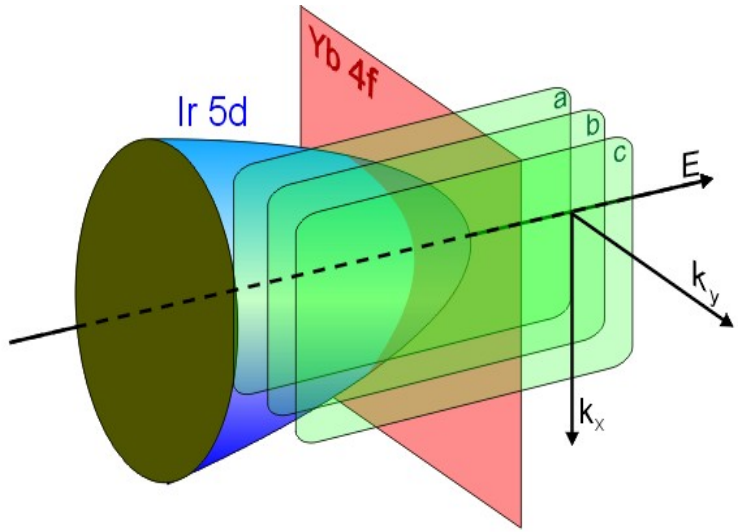


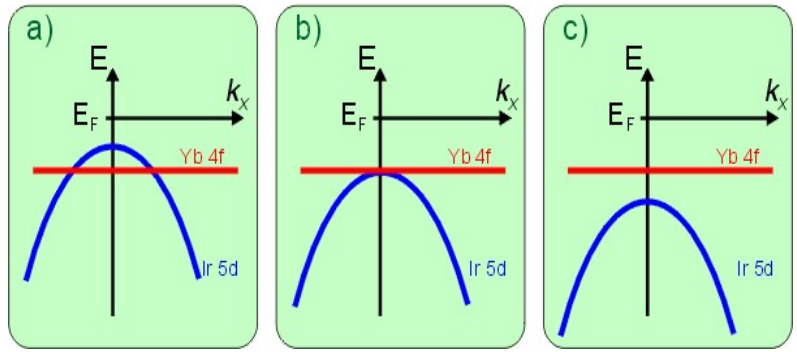


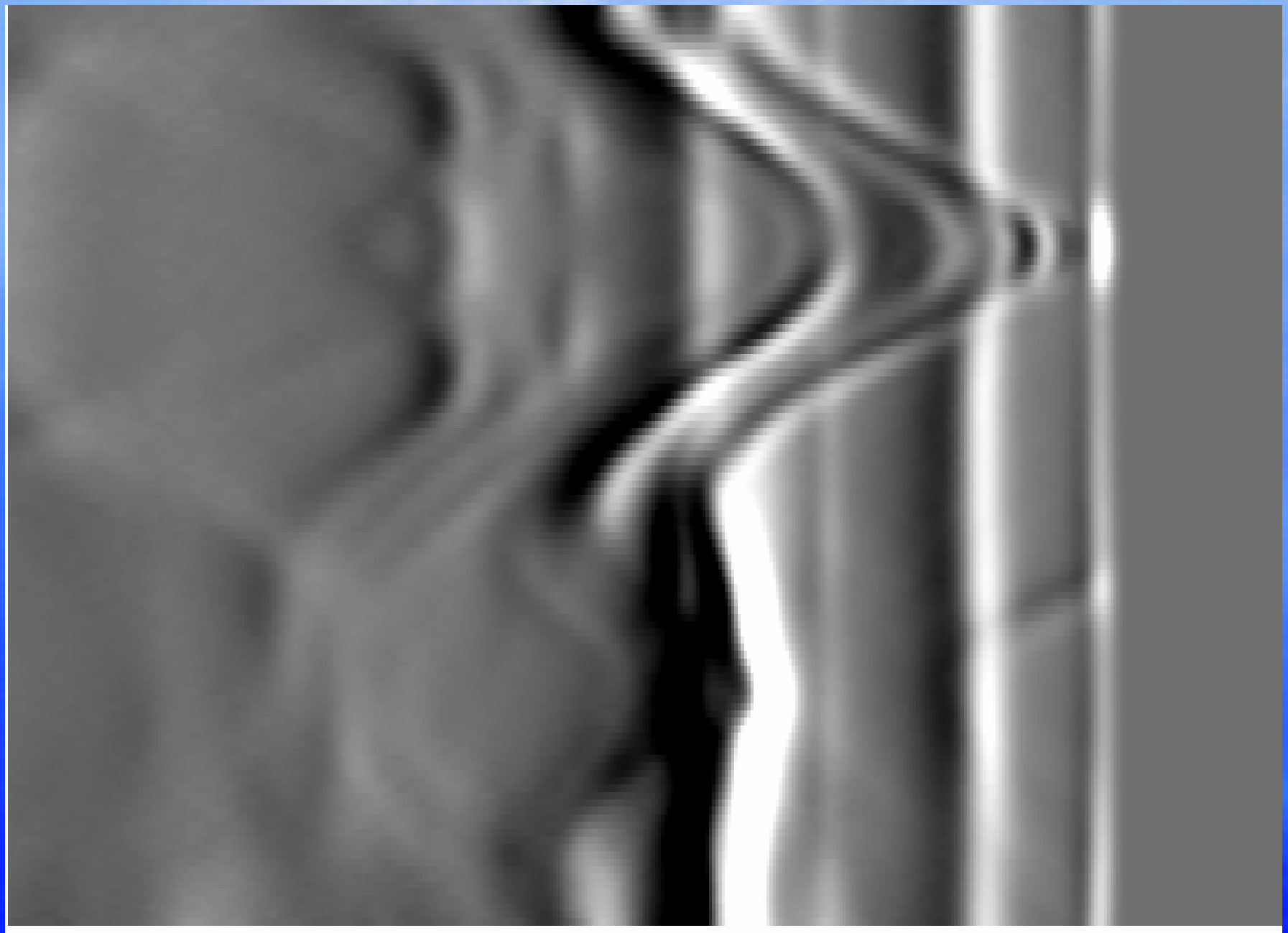

YbIr2Si2
hν = 110 eV
bulk valence: 2,92
surface valence : 2
Yb3+ (bulk)
Yb2+ (bulk)
Yb2+ (surf.)
Intensity
12
10
8
6
4
2
EF
Binding Energy (eV)

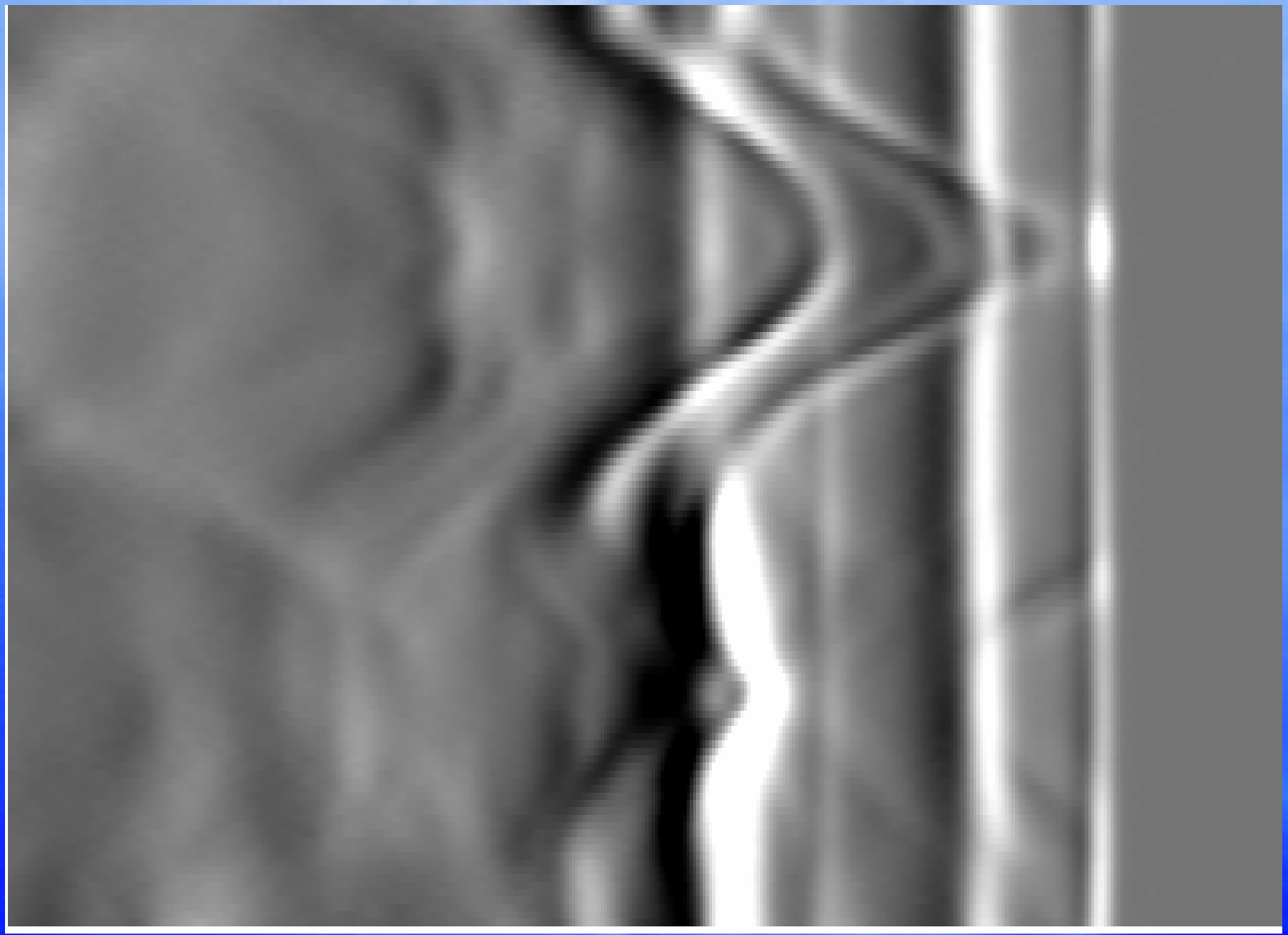
k-dependent hybridization in $YbIr_2Si_2$

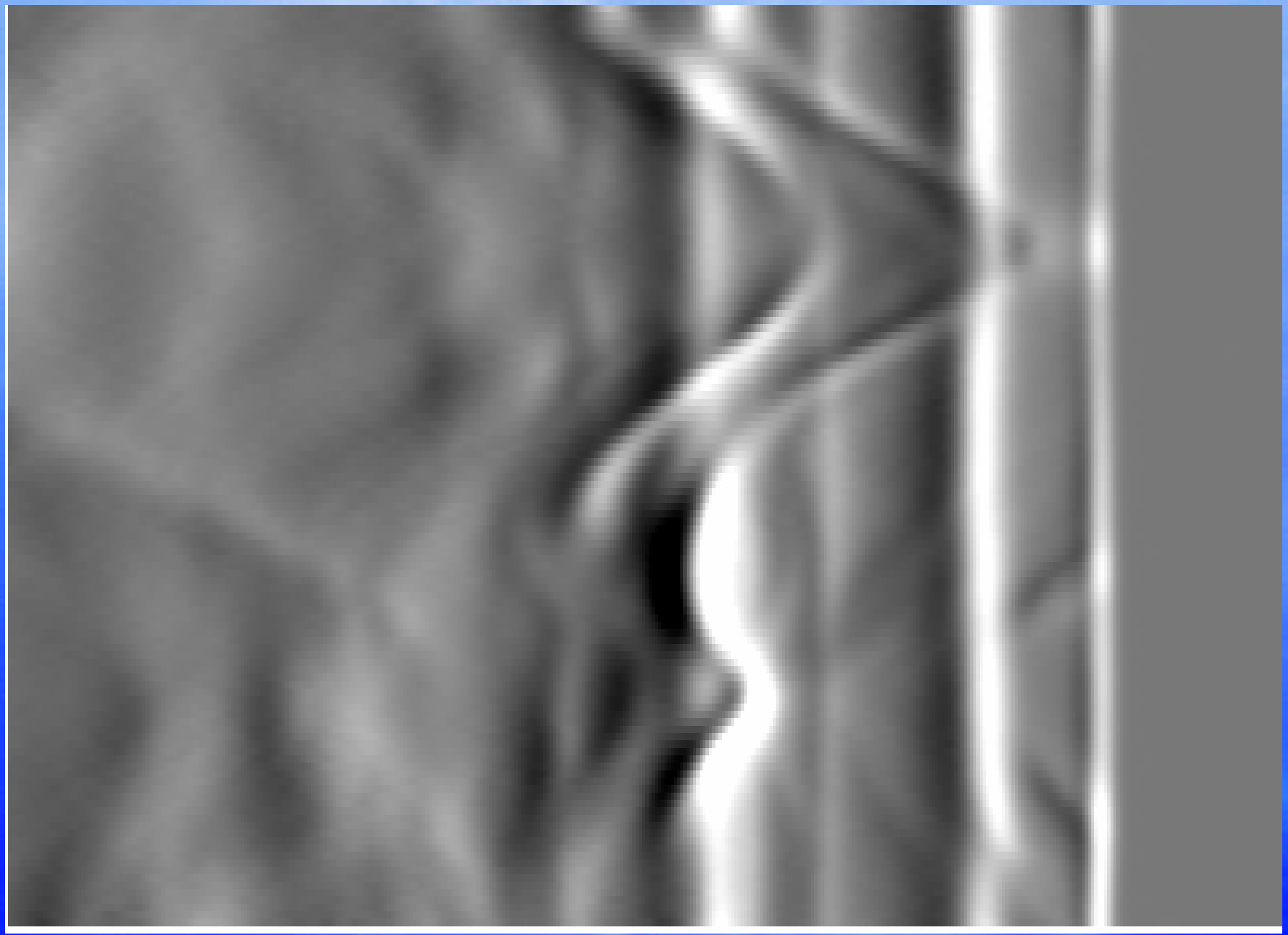


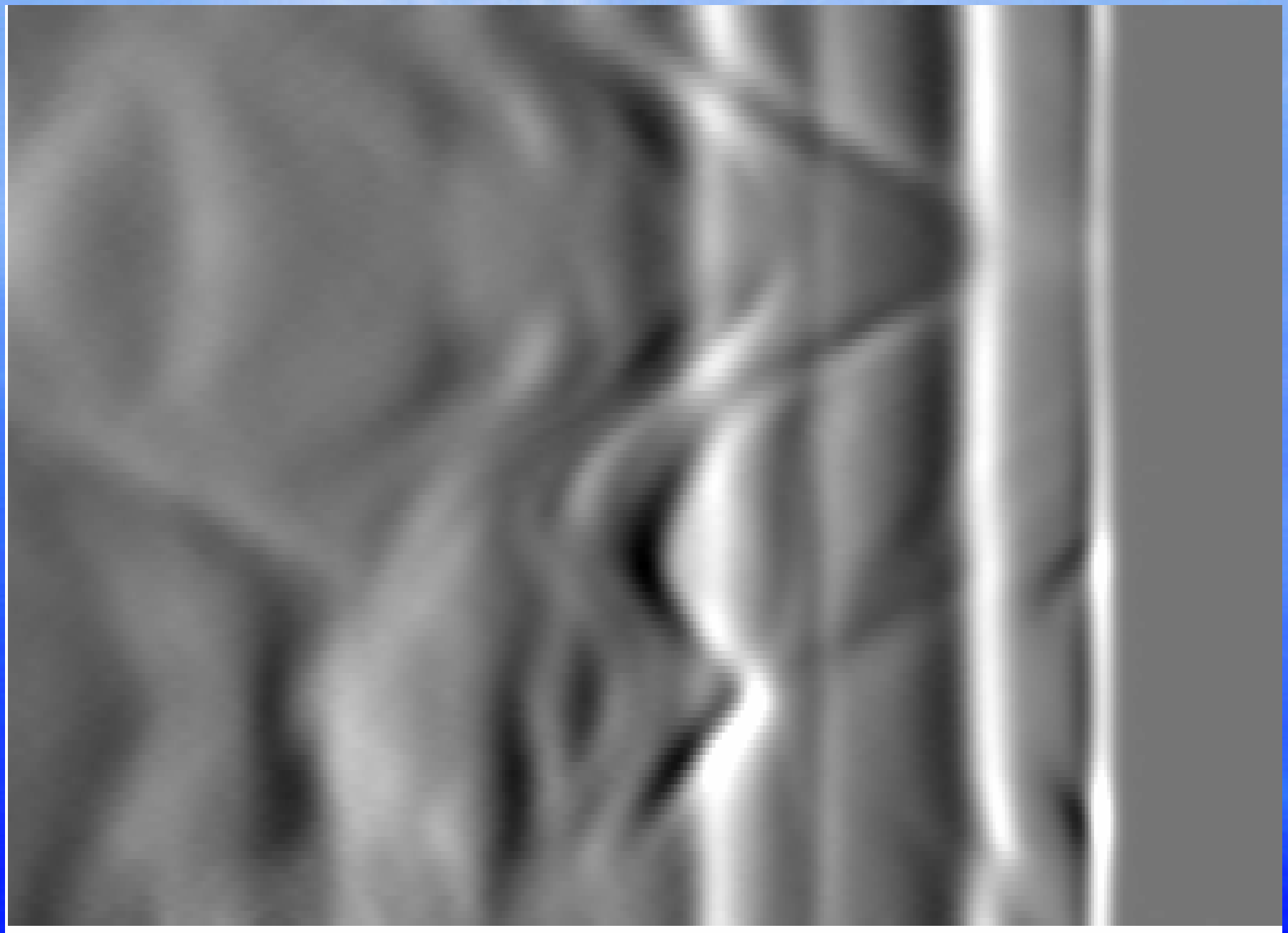


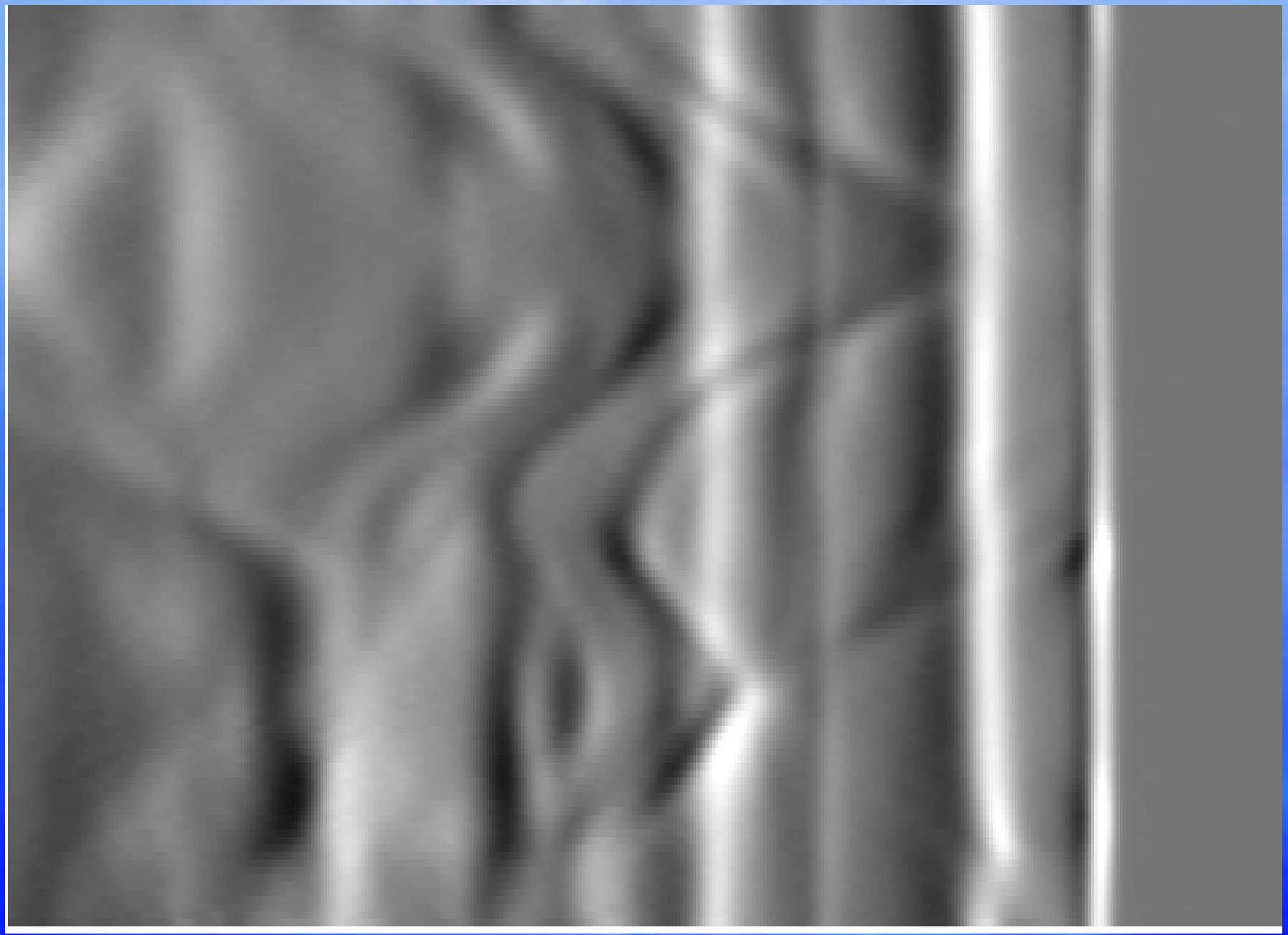


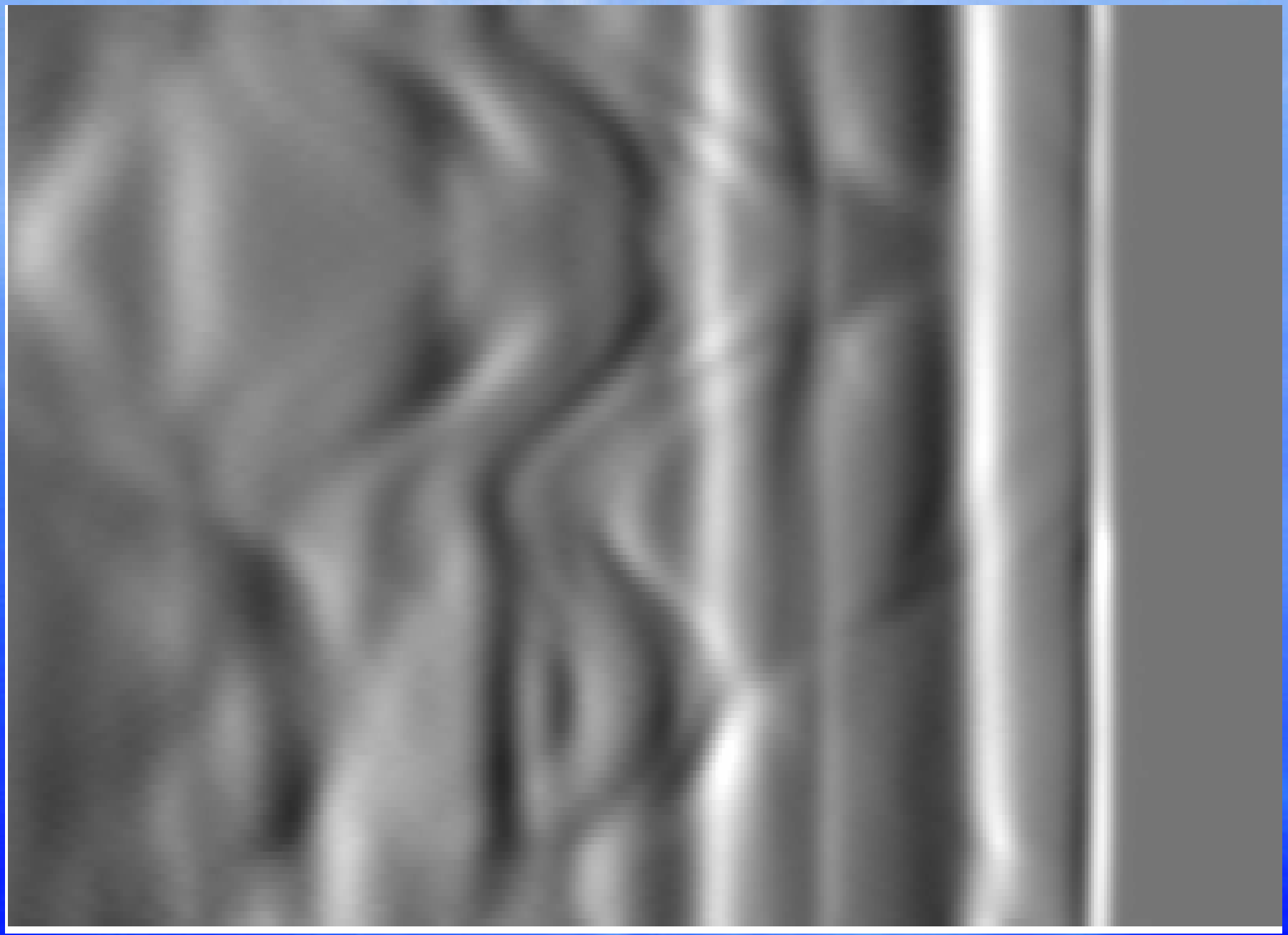


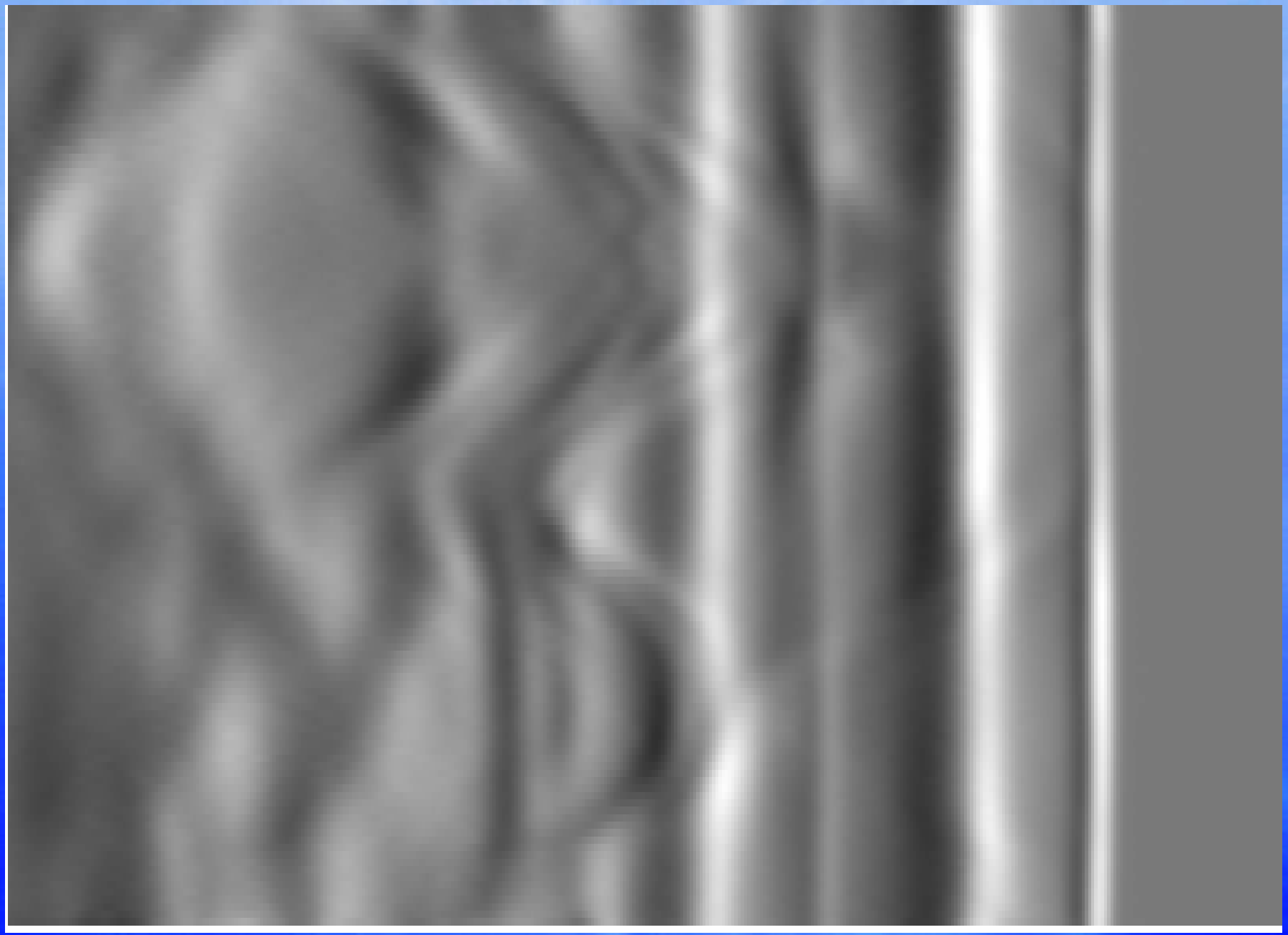


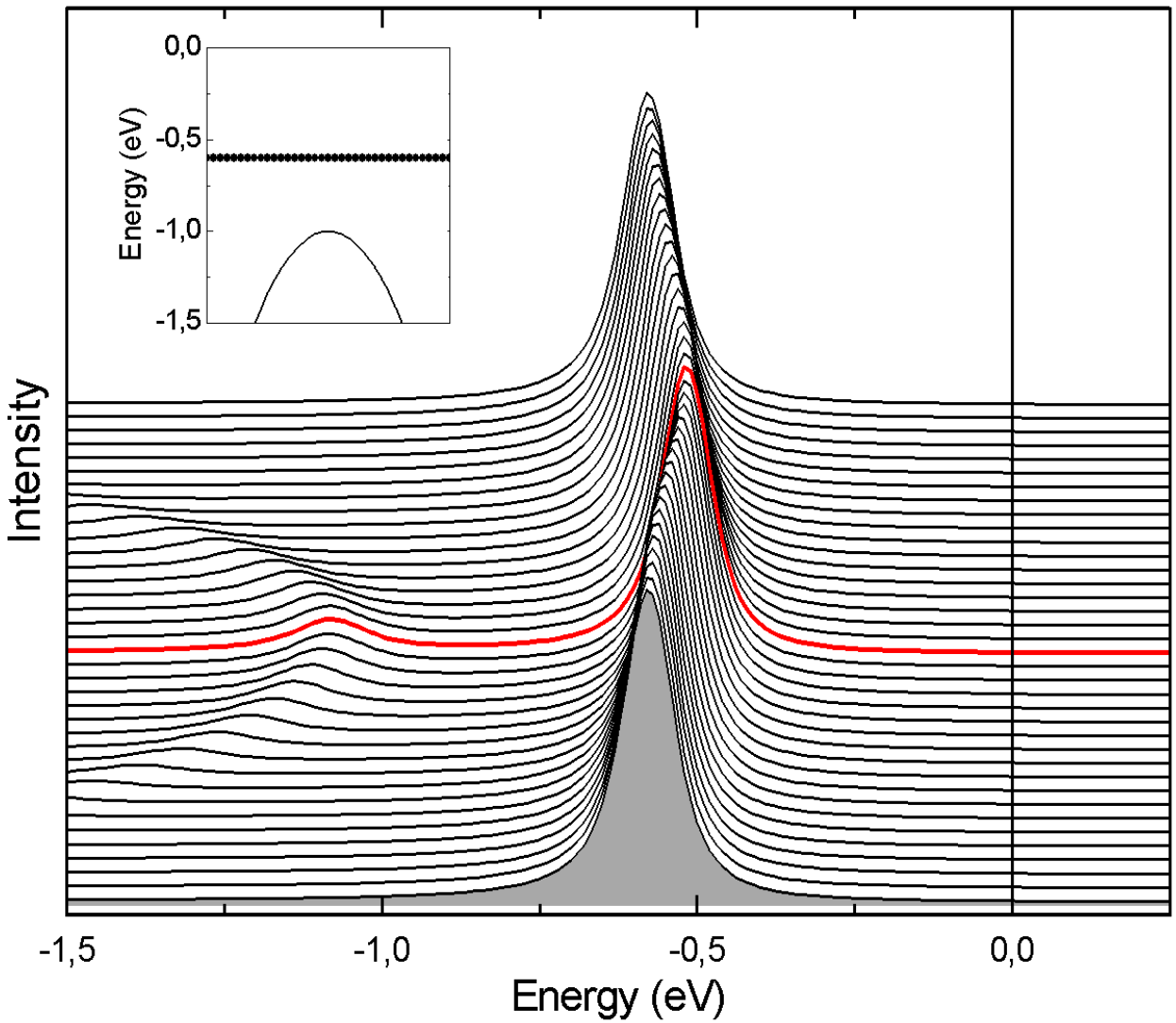


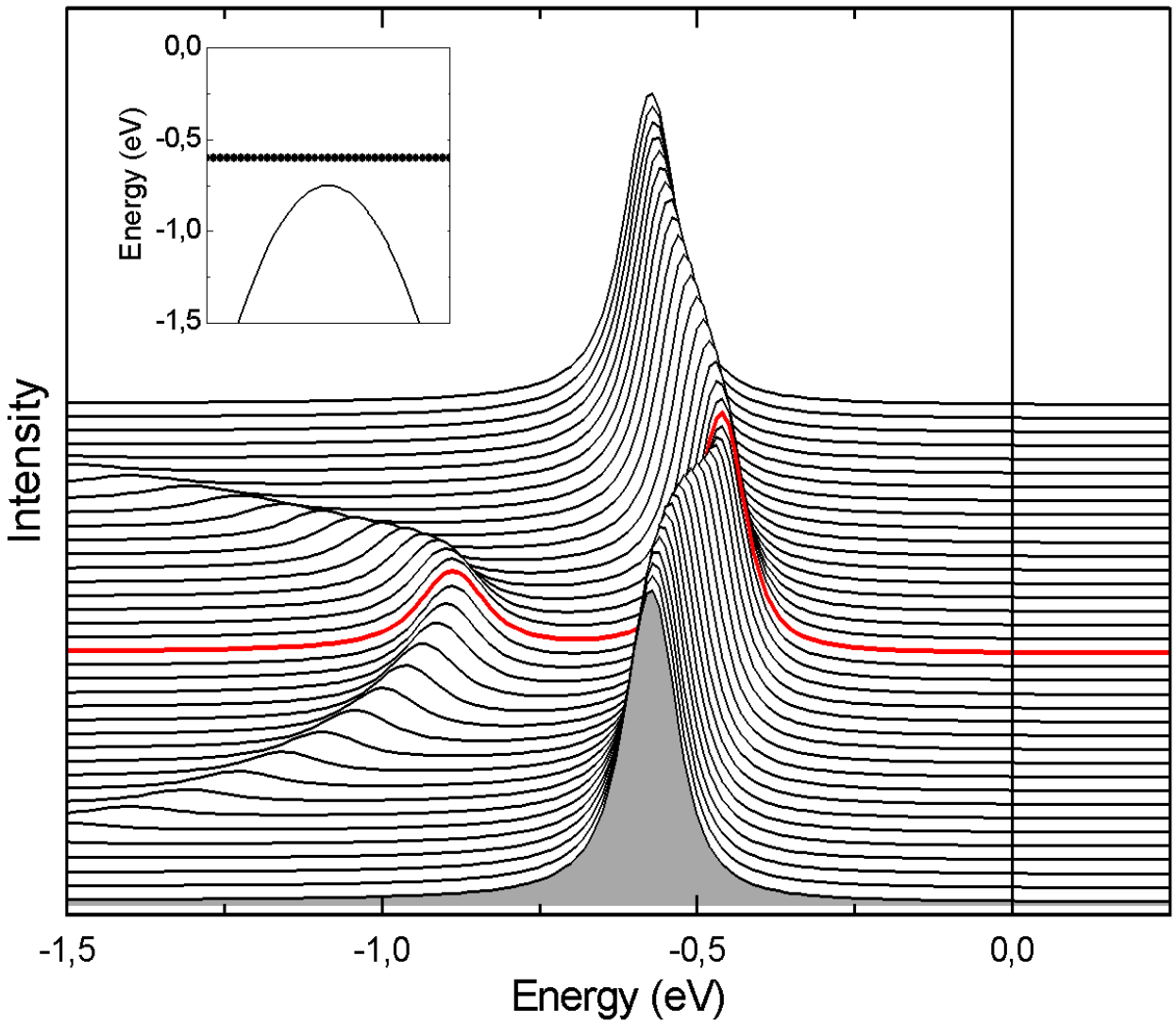


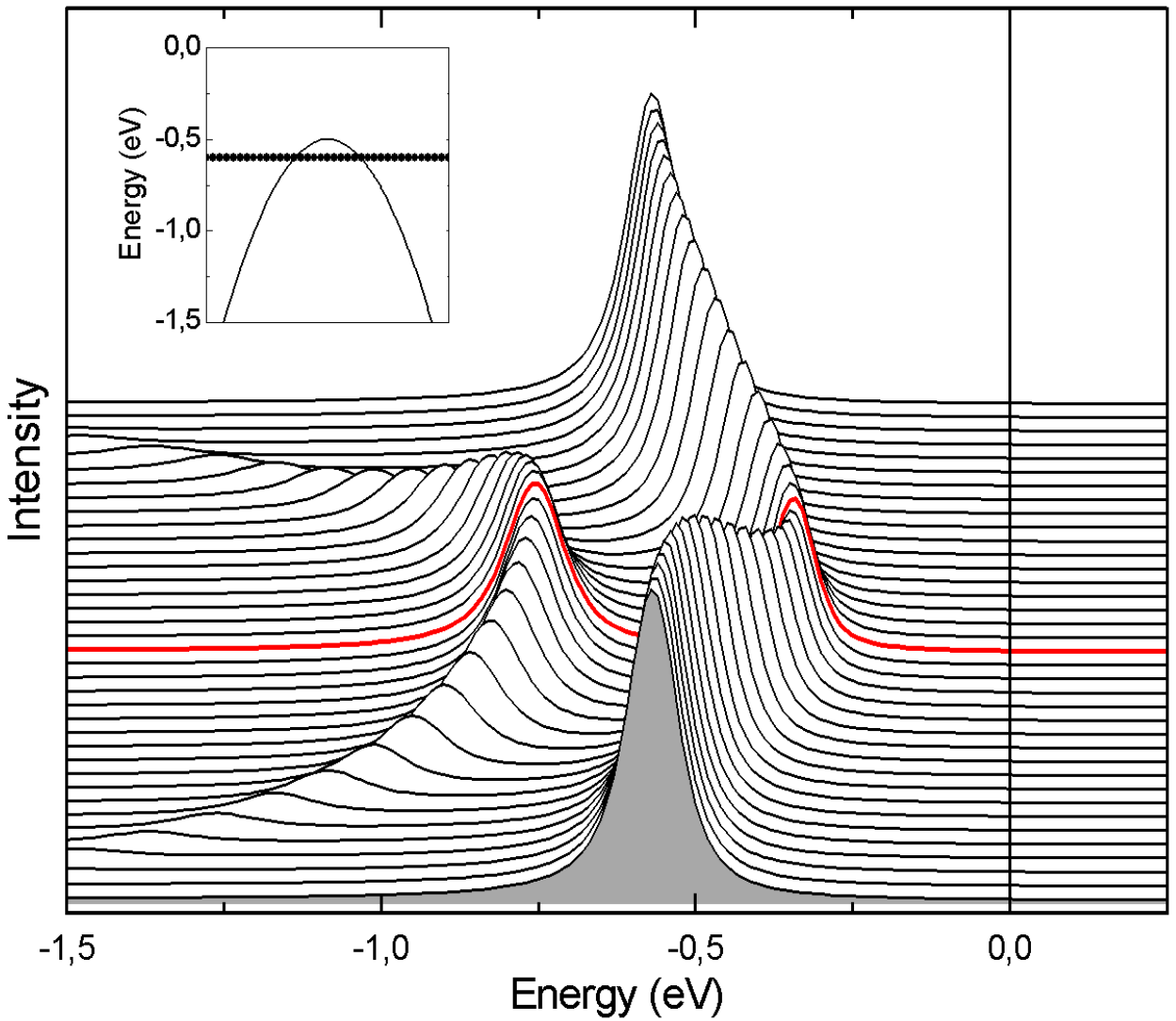


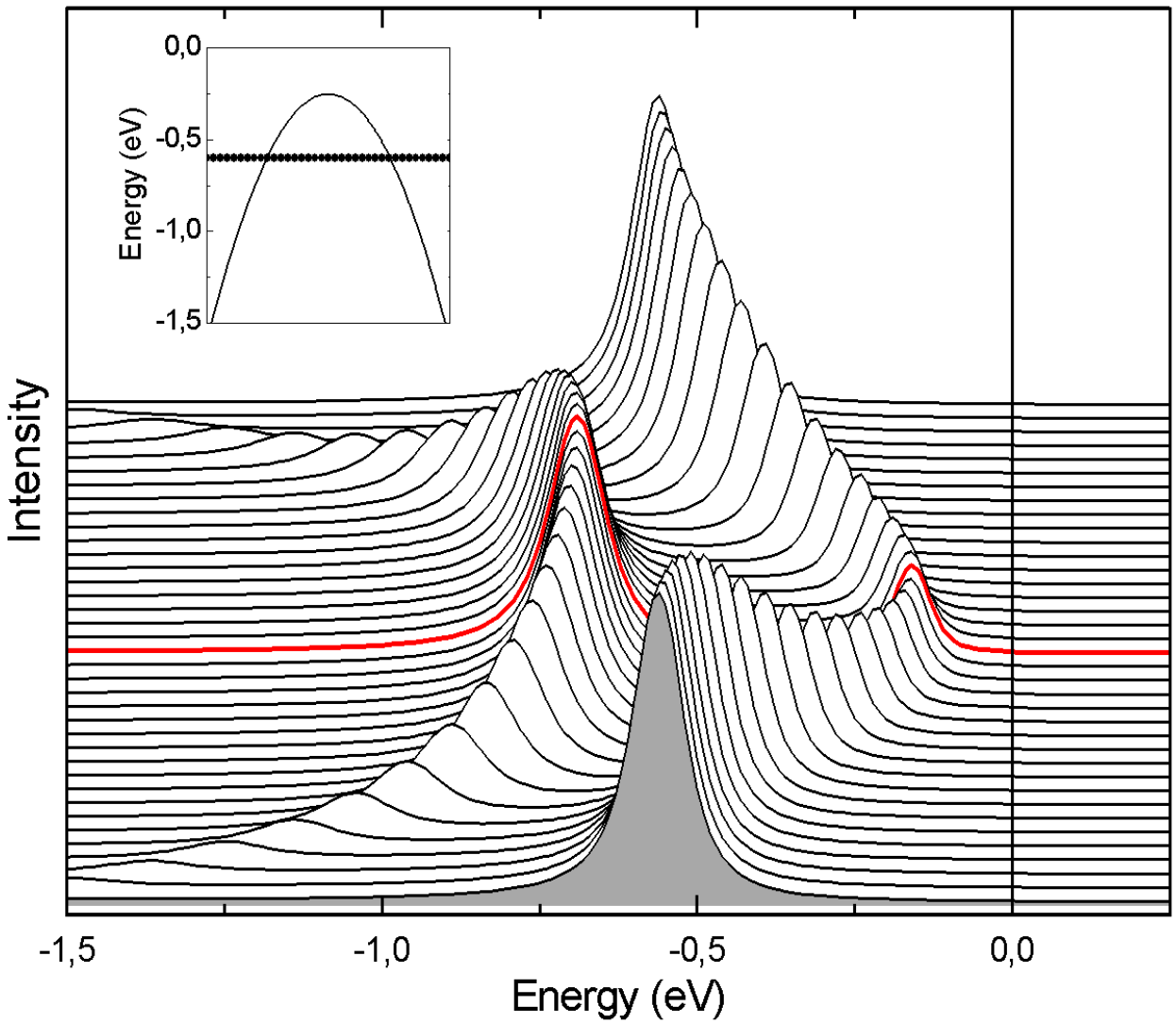


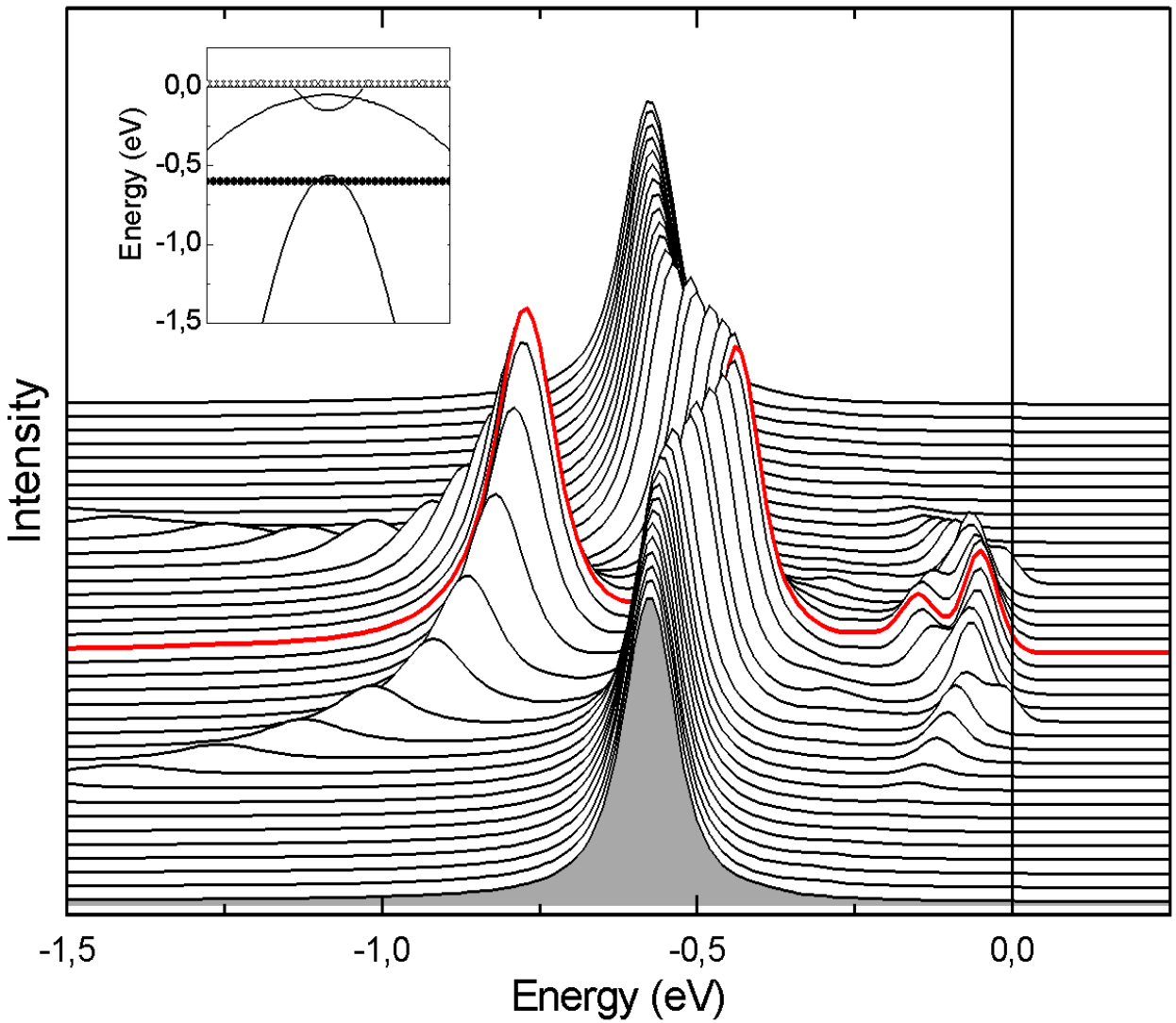


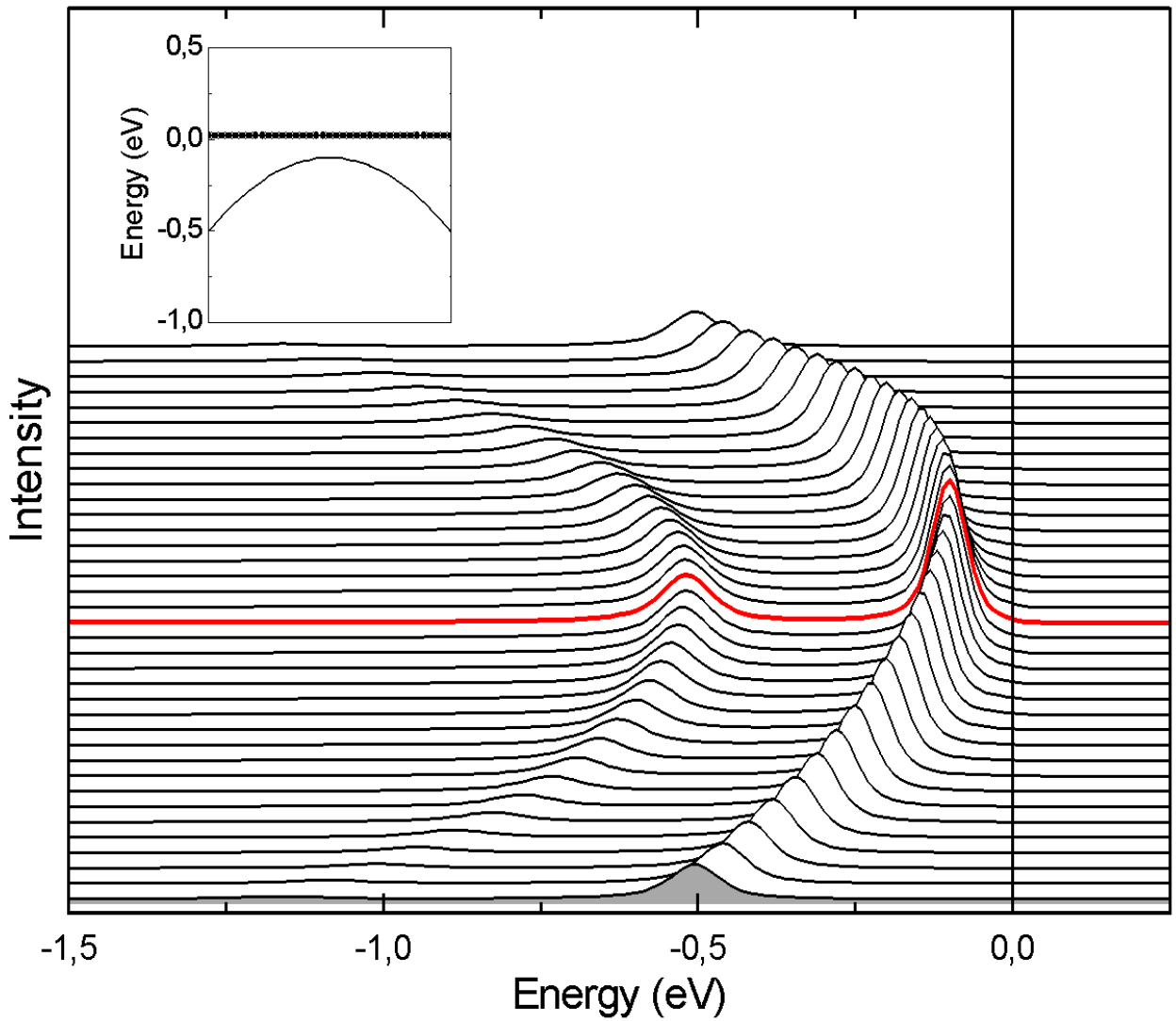

0,0
-0,5
-1,0
-1,5
Energy (eV)
Intensity
-1,5
-1,0
-0,5
0,0
Energy (eV)

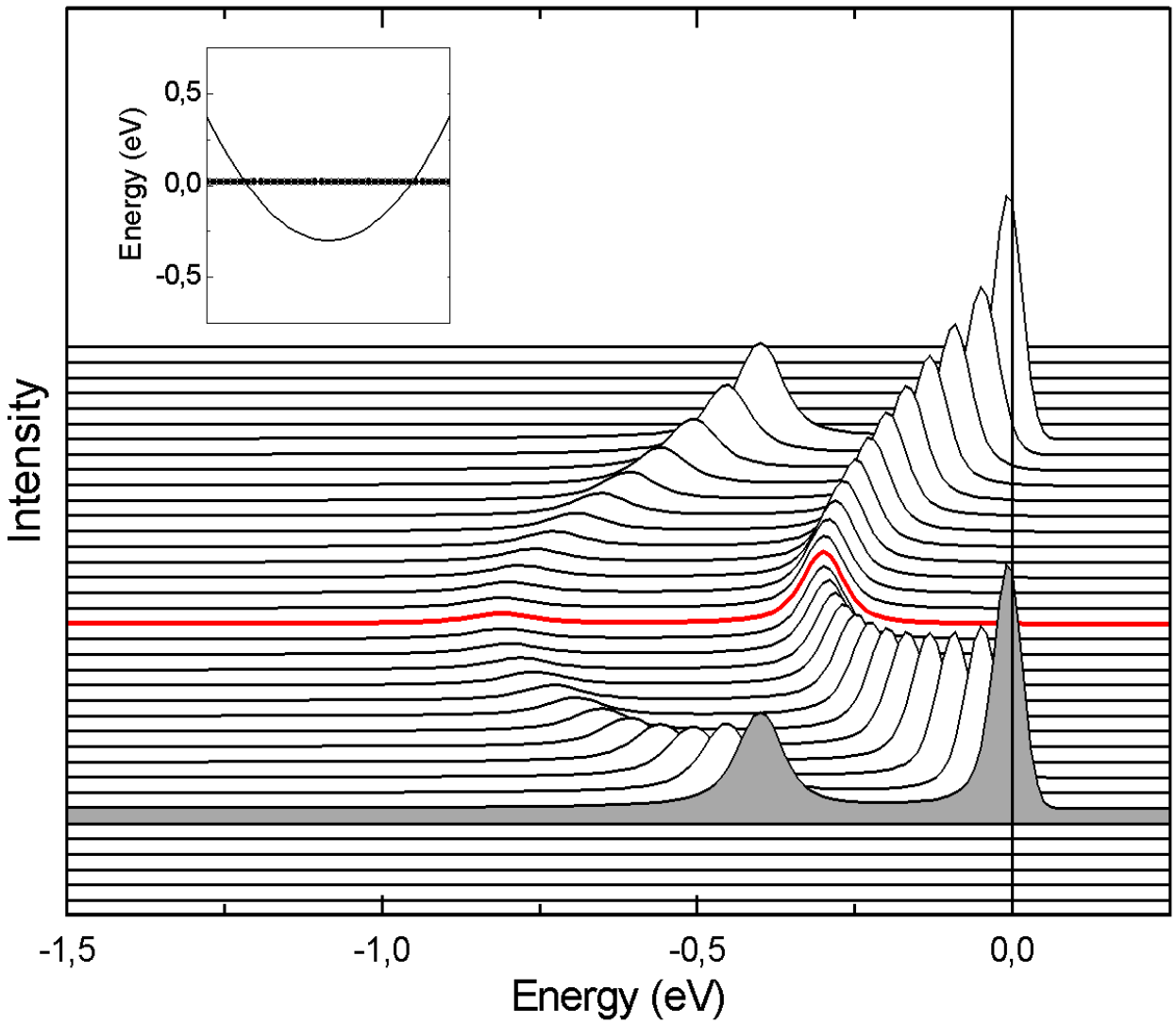

0,0
-0,5
-1,0
-1,5
Energy (eV)
Intensity
-1,5
-1,0
-0,5
0,0
Energy (eV)

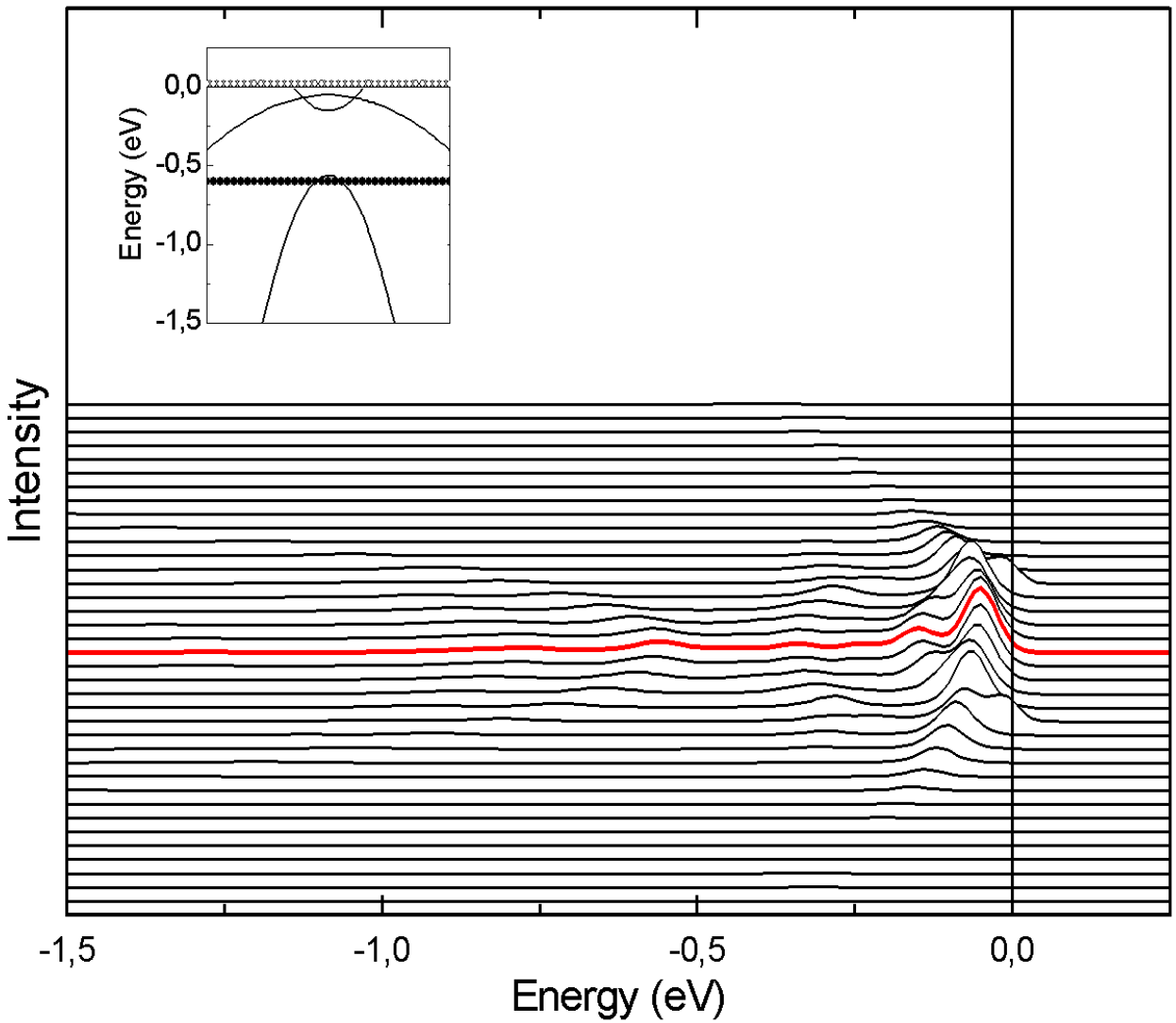

0,0
-0,5
-1,0
-1,5
Energy (eV)
Intensity
-1,5
-1,0
-0,5
0,0
Energy (eV)

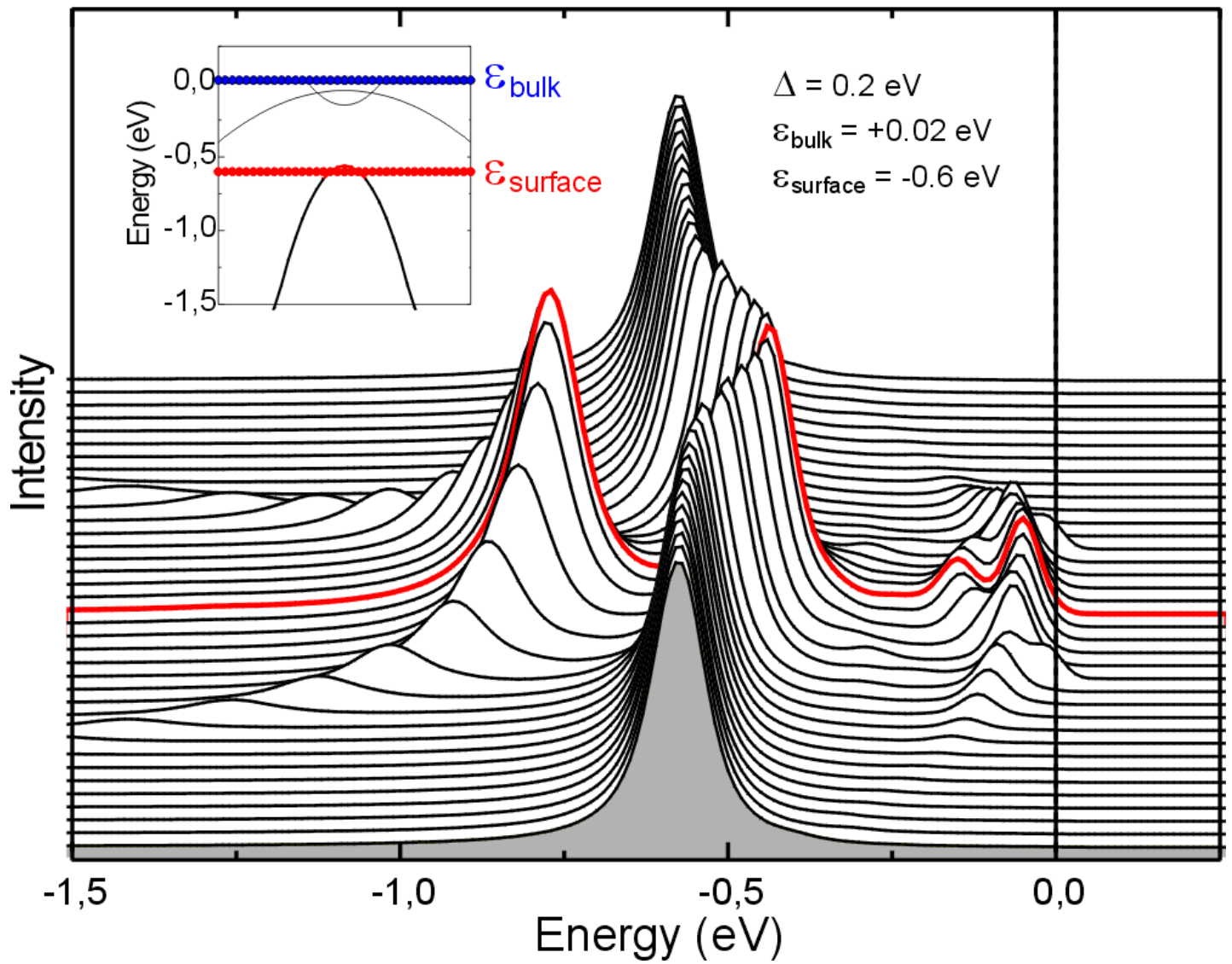

0,0
-0,5
-1,0
-1,5
Energy (eV)
Intensity
-1,5
-1,0
-0,5
0,0
Energy (eV)

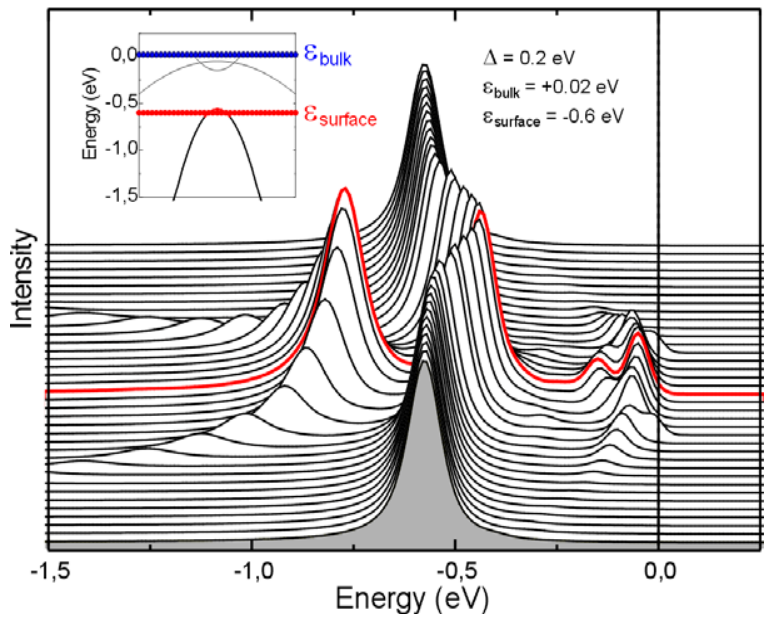

Intensity
Energy (eV)
-1,5
-1,0
-0,5
0,0
Energy (eV)
0,0
-0,5
-1,0
-1,5


0,5
0,0
-0,5
-1,0
Energy (eV)
Intensity
-1,5
-1,0
-0,5
0,0
Energy (eV)

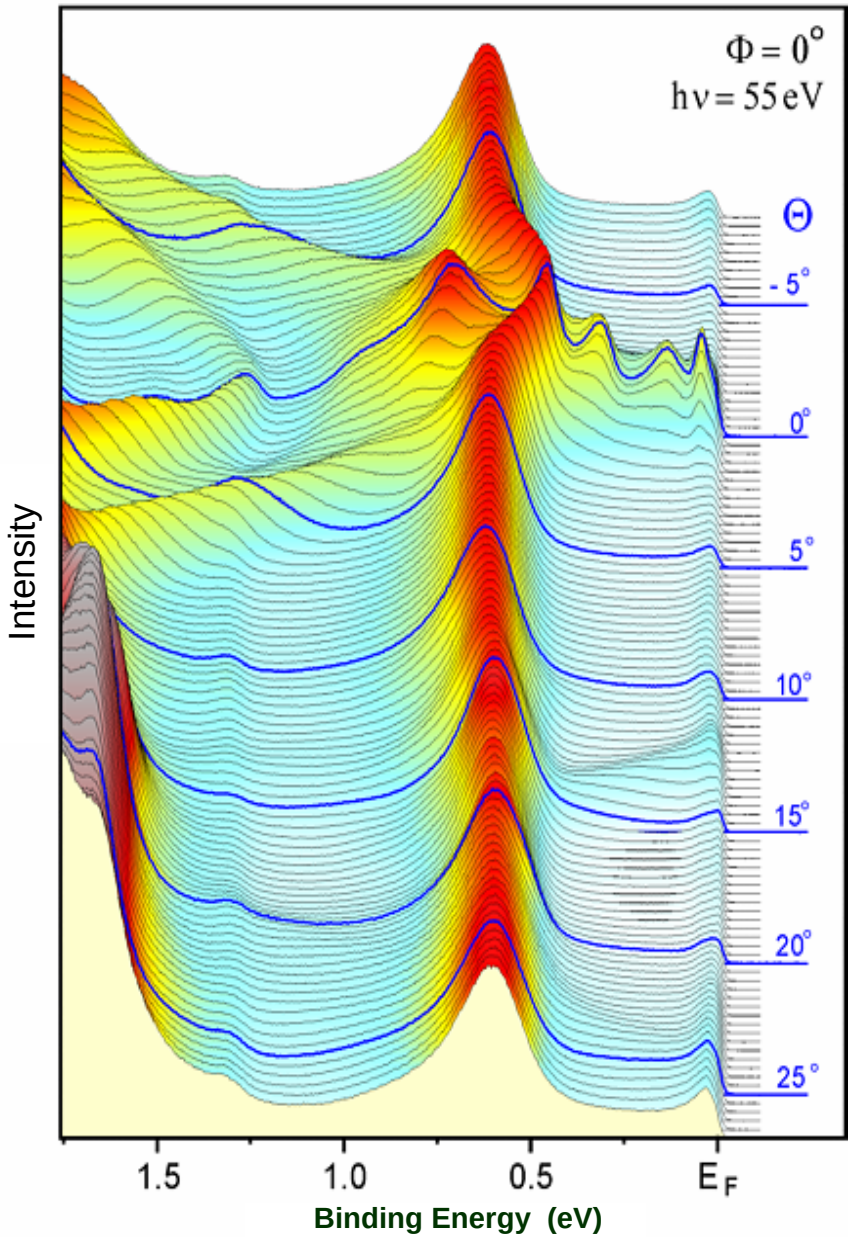

Energy (eV)
0,5
0,0
-0,5
Intensity
-1,5
-1,0
-0,5
0,0
Energy (eV)


Intensity
Energy (eV)
-1,5
-1,0
-0,5
0,0
Energy (eV)
0,0
-0,5
-1,0
-1,5


Δ = 0.2 eV
ε bulk = +0.02 eV
ε surface = -0.6 eV
ε bulk
ε surface
Energy (eV)
0,0
-0,5
-1,0
-1,5
Intensity
-1,5
-1,0
-0,5
0,0
Energy (eV)



- Good agreement between theory and experiment!
- Fermi-level peak far away from Γ may be caused by:
  i) thermal excitations
  ii) interactions of unoccupied f and VB states
  iii) partial integration over k due to finite U_{ff}



Summary

- **In the limit of negligible hybridization 4f states behave as core-levels.**
- **Hybridization effects are usually handled in the light of the Single Impurity Anderson Model (SIAM).**
- **Angle-resolved photoemission data show that hybridization depends on and varies with wave-vector.**
- **Data analysis is possible within a simple approach to the Periodic Anderson Model (PAM) that has the form of SIAM but with direction dependent hybridization.**
- **Similar direction dependent approaches may explain anisotropies of other physical properties of mixed-valence and heavy-fermion systems.**

Collaboration:

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MPI cPfS:

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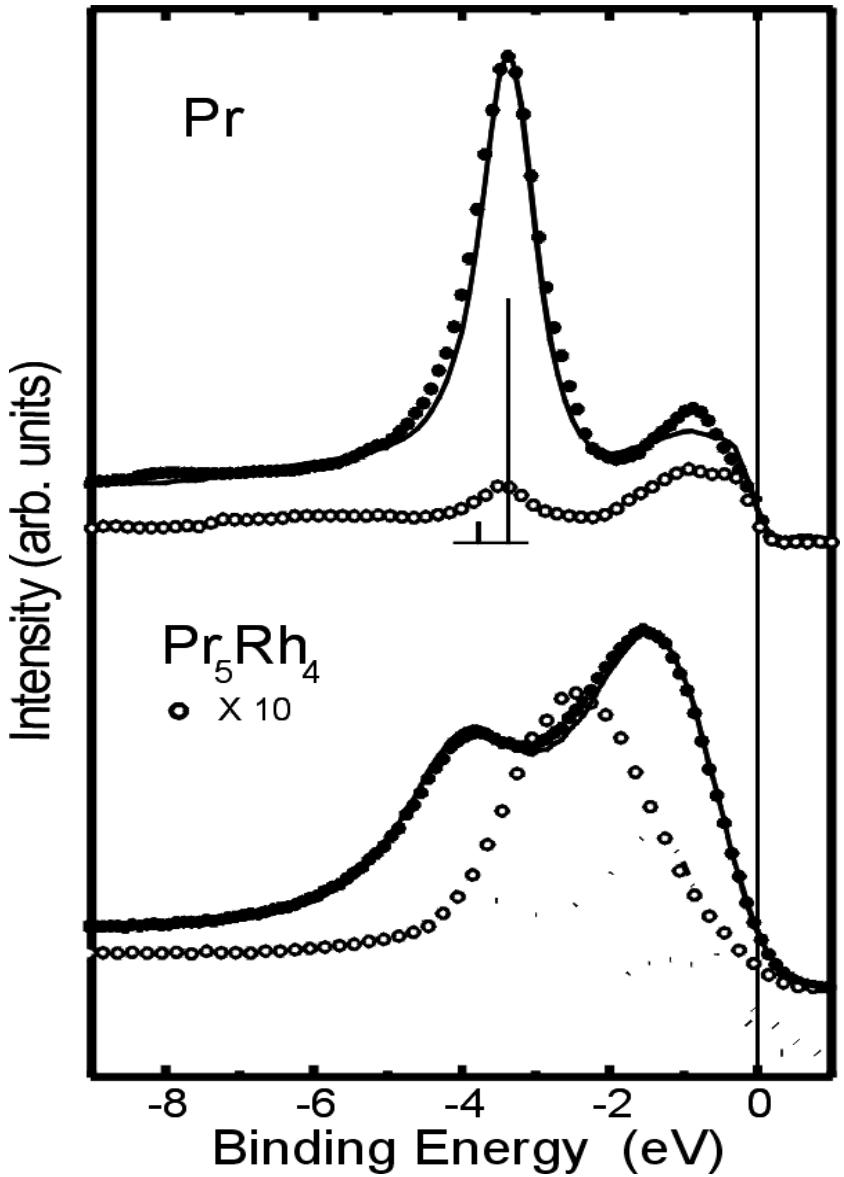
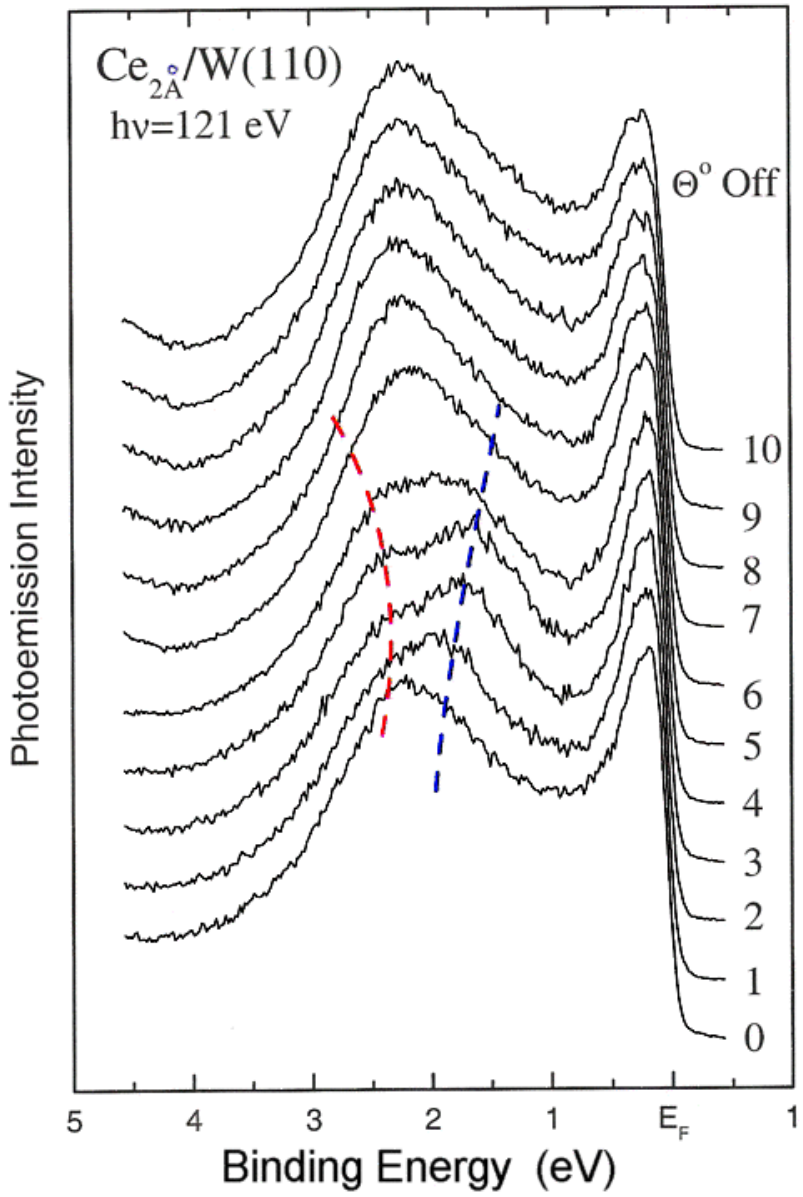
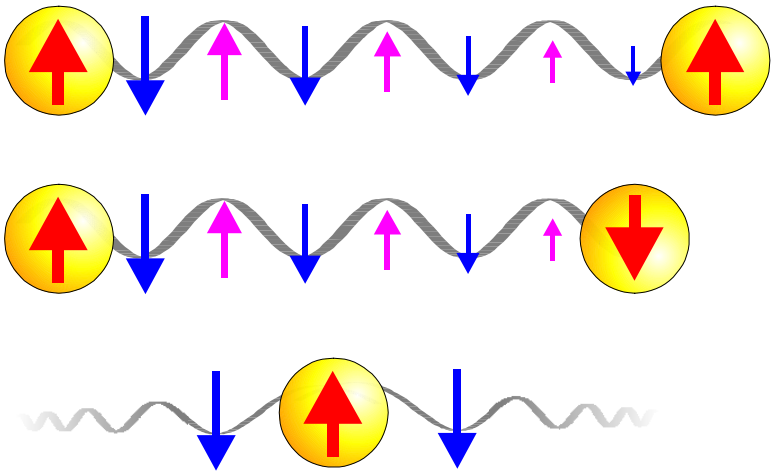

Pr
Pr_5Rh_4
X 10
Intensity (arb. units)
-8
-6
-4
-2
0
Binding Energy (eV)


$Ce_{2Å}$/W(110)
hν=121 eV
Θ° Off
10
9
8
7
6
5
4
3
2
1
0
Photoemission Intensity
5
4
3
2
1
E_F
1
Binding Energy (eV)

Interactions of localized moments:

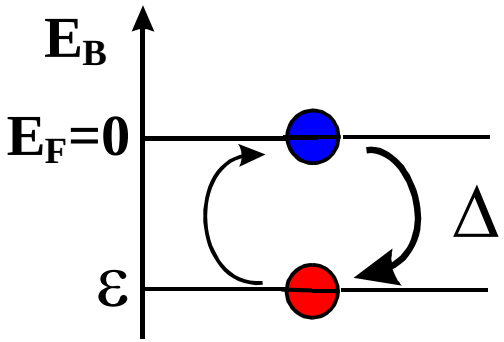


RKKY interaction:

oscillatoric spin polarization
of conduction electrons
→ magnetic order

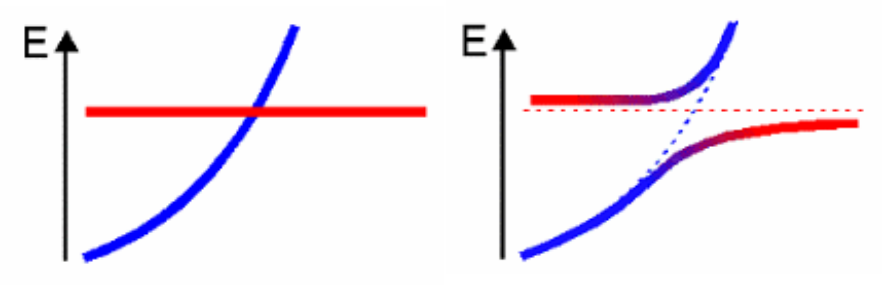
Kondo-effect:

screening of local moment
→ increase of resistivity below T_K
→ large density of states at E_F
→ large specific heat



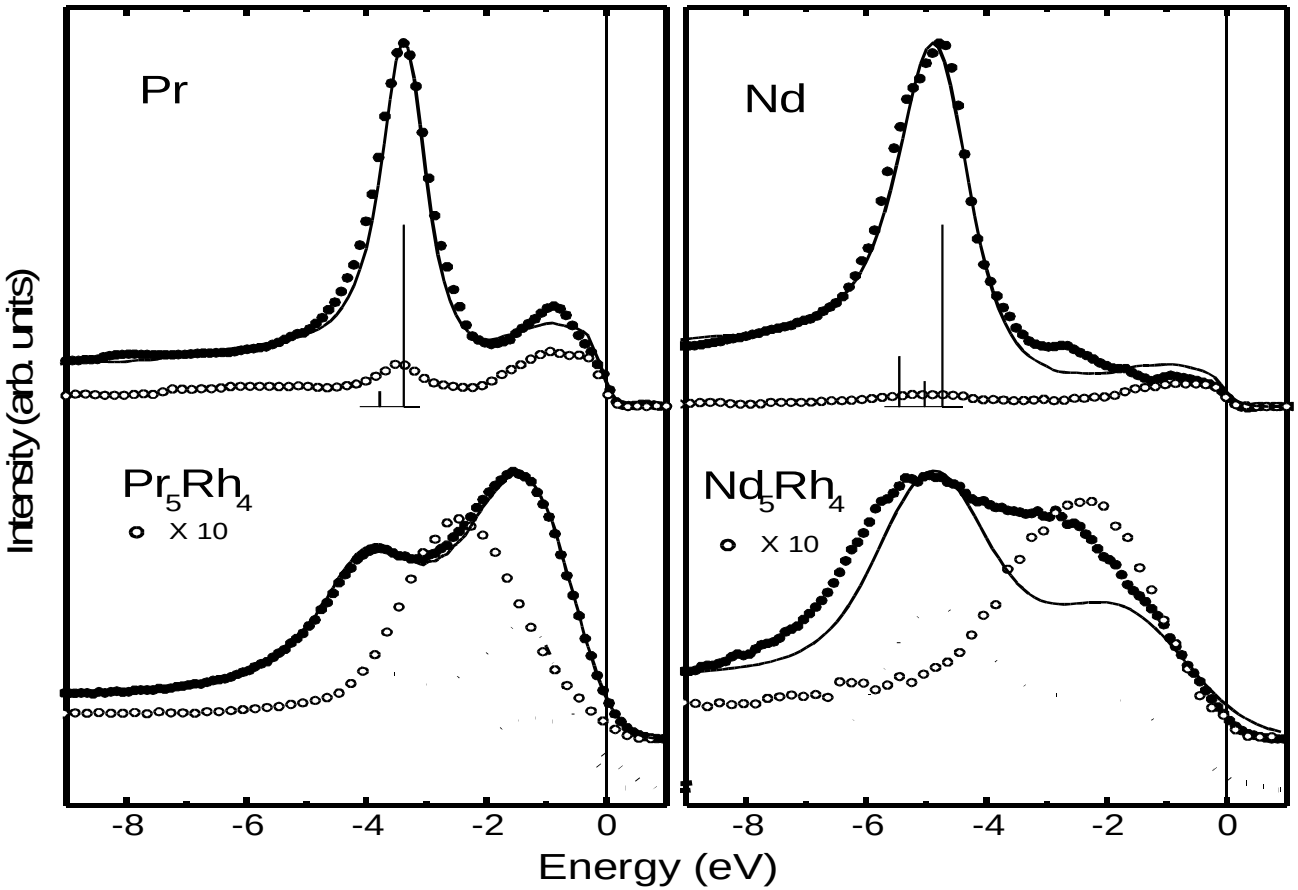
Anderson model:

hopping between localized
and itinerant states



→ **heavy fermion behavior**

f-d interaction in Pr and Nd systems



Laubschat *et al.*, J. Electr. Spectr. Rel. Phen. 128 (2003) 45