e-e interactions for strongly -and not so strongly- correlated materials

Outline:

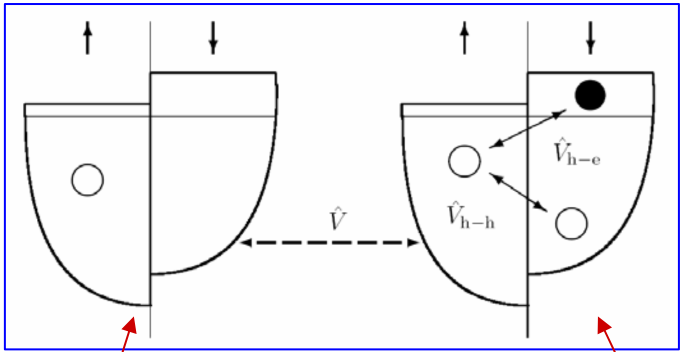
- *Method to solve HH*
- *Charged excitations – photoemission from valence and core states*
- *Neutral excitations – XAS and XMCD*

Franca Manghi

$$\hat{H} = \sum_{i\alpha\sigma} \epsilon_{i\alpha\sigma} \hat{n}_{i\alpha\sigma} \quad + \quad \sum_{i\alpha\beta\sigma} t_{i\alpha\beta} \hat{c}^{\dagger}_{i\alpha\sigma} \hat{c}_{j\beta\sigma}$$

$$+ \quad U \quad \sum_{i\alpha\beta\sigma} \hat{n}_{i\alpha\sigma} \hat{n}_{i\beta-\sigma} + (U - J) \sum_{i\alpha\beta\sigma} \hat{n}_{i\alpha\sigma} \hat{n}_{i\beta\sigma}$$

3-body scattering



$$\hat{H} = \sum_{mm'} < m|\hat{H}|m'> \cdot |m><m'| = \hat{H}_1 + \hat{H}_3 + \hat{V}$$

$$|u> \equiv \hat{c}_k|0> \qquad |t> \equiv \hat{c}^{\dagger}_{q_3}\hat{c}_{q_2}\hat{c}_{q_1}|0>$$

$$G(k,\omega) = <\Psi_k^{N-1}|\frac{1}{z-\hat{H}}|\Psi_k^{N-1}> \qquad \boxed{\simeq <u|\frac{1}{z-\hat{H}}|u>}$$

$$G(k,\omega) = -\frac{1}{\omega - \epsilon_k + \sum_{tt'} F^3_{tt'} V_{t'u} V_{ut'}}$$

$$\Sigma(\omega)$$

$$\boxed{\cancel{q_1 + q_2 - q_3 = k}}$$

3BS chronology

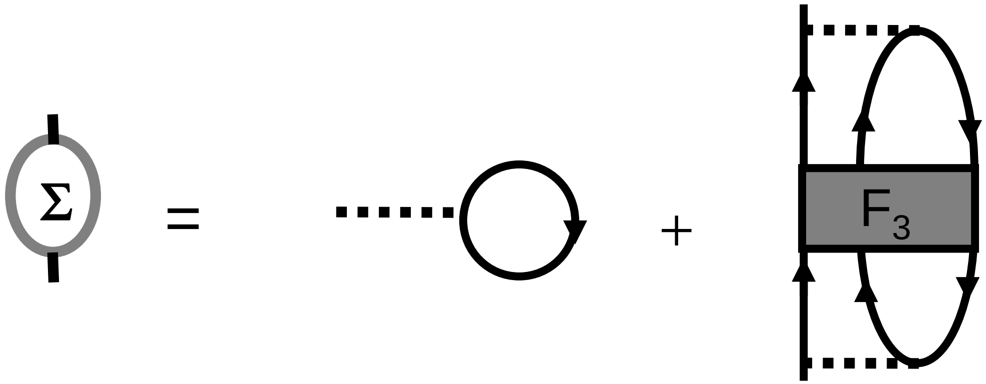
1983: born in Japan Igarashi J. Phys. Soc. Jpn.52, 2827 (1983)

90s: developed in Modena

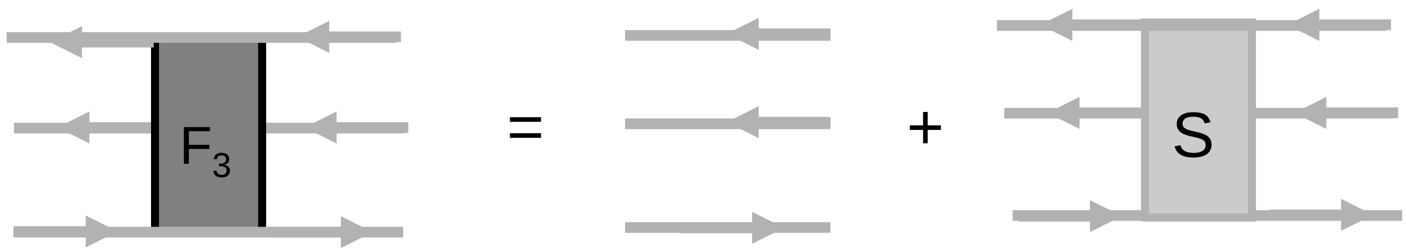
NiO PRL 73 3129 (1994)

Ni: PRB 56 7149 (1997) …

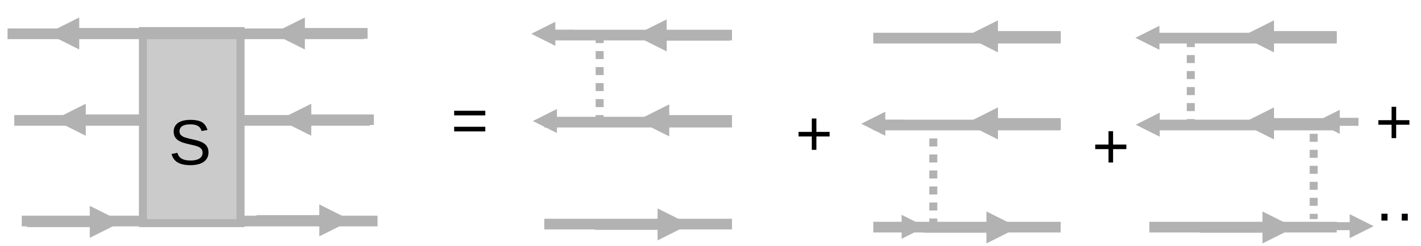
Diamagratically:



where



and



self-energy is the result of multiple h-h and e-h scatterings

In practice:

- Input: band structure $\boxed{(\epsilon_k^n, C_{\alpha\uparrow}^n(k), n_{\alpha\uparrow}(\epsilon)) \text{ and } U;}$
- free propagators

$$g_{h-h}^{\alpha\beta}(\omega) = \int_{-\infty}^{E_f} \mathrm{d}\,\epsilon' \int_{-\infty}^{E_f} \mathrm{d}\,\epsilon \frac{n_{\alpha\downarrow}(\epsilon) n_{\beta\uparrow}(\epsilon')}{\omega - \epsilon' - \epsilon - i\delta},$$

$$g_{h-e}^{\alpha\beta}(\omega) = \int_{-\infty}^{E_f} \mathrm{d}\,\epsilon' \int_{E_f}^{\infty} \mathrm{d}\,\epsilon \frac{n_{\alpha\downarrow}(\epsilon) n_{\beta\uparrow}(\epsilon')}{\omega - \epsilon' + \epsilon - i\delta}, \qquad g^{\beta}(\omega) = \int_{-\infty}^{E_f} \mathrm{d}\,\epsilon' \frac{n_{\beta\uparrow}(\epsilon')}{\omega - \epsilon' - i\delta};$$

- T-matrices

$$T_{h-h}^{\alpha\beta}(\omega) = \frac{U}{1 + U g_{h-h}^{\alpha\beta}(\omega)}, \qquad T_{h-e}^{\alpha\beta}(\omega) = \frac{-U}{1 - U g_{h-e}^{\alpha\beta}(\omega)};$$

- kernel

$$K^{\alpha\beta}(\omega, \epsilon, \epsilon') = \int_{-\infty}^{E_f} \mathrm{d}\,\epsilon'' n_{\alpha\downarrow}(\epsilon'') g^{\beta}(\omega + \epsilon'' - \epsilon) g^{\beta}(\omega + \epsilon'' - \epsilon') T_{h-e}^{\alpha\beta}(\omega + \epsilon'') T_{h-h}^{\alpha\beta}(\omega - \epsilon''),$$

and

$$\begin{aligned} B^{\alpha\beta}(\omega, \epsilon) &= \int_{-\infty}^{E_f} \mathrm{d}\,\epsilon' n_{\alpha\downarrow}(\epsilon') g^{\beta}(\omega + \epsilon' - \epsilon) T_{h-e}^{\alpha\beta}(\omega + \epsilon') \\ &\times \left[g_{h-e}^{\alpha\beta}(\omega - \epsilon') + \int_{E_f}^{\infty} \mathrm{d}\,\epsilon'' n_{\alpha\downarrow}(\epsilon'') g^{\beta}(\omega + \epsilon' - \epsilon'') g_{h-h}^{\alpha\beta}(\omega - \epsilon'') T_{h-h}^{\alpha\beta}(\omega - \epsilon'') \right]; \end{aligned}$$

In practice:

- solve the integral equation

$$A^{\alpha\beta}(\omega,\epsilon) = B^{\alpha\beta}(\omega,\epsilon) + \int_{E_f}^{\infty} \mathrm{d}\,\epsilon' n_{\alpha\downarrow}(\epsilon') K^{\alpha\beta}(\omega,\epsilon,\epsilon') A^{\alpha\beta}(\omega,\epsilon');$$

- orbital self-energy

$$\Sigma^{-}_{\beta\uparrow}(\omega) = \sum_{\alpha} \int_{E_f}^{\infty} \mathrm{d}\,\epsilon\, n_{\alpha\downarrow}(\epsilon) T^{\alpha\beta}_{h-h}(\omega-\epsilon) \left[1 + U A^{\alpha\beta}(\omega\epsilon)\right];$$

- $\mathbf{K}$- and band-index dependent self-energy

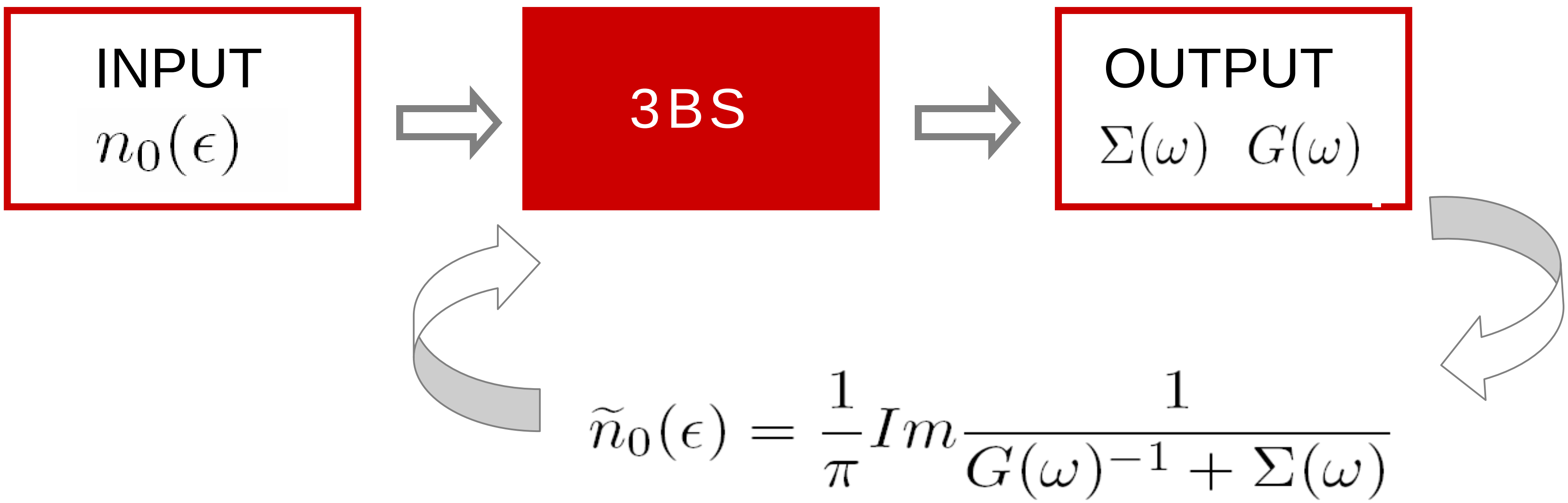
$$\Sigma^{-}(kn\uparrow,\omega) = U \sum_{\beta} |C^{n}_{\beta\uparrow}(k)|^2 \left[\sum_{\alpha} \int_{-\infty}^{E_f} \mathrm{d}\,\epsilon\, n_{\alpha\downarrow}(\epsilon) \Sigma^{-}_{\beta\uparrow}(\omega)\right];$$

•For ***small*** U analitically recovers 2nd order perutbation theory

•For ***infinite*** U reproduces the atomic limit

•Kanamori T-Matrix approach is its limit for almost fully occupied band

For intermediate U:

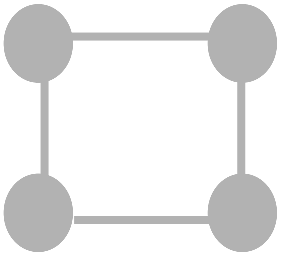


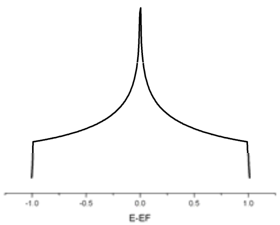
$$\tilde{n}_0(\epsilon) = \frac{1}{\pi} Im \frac{1}{G(\omega)^{-1} + \Sigma(\omega)}$$

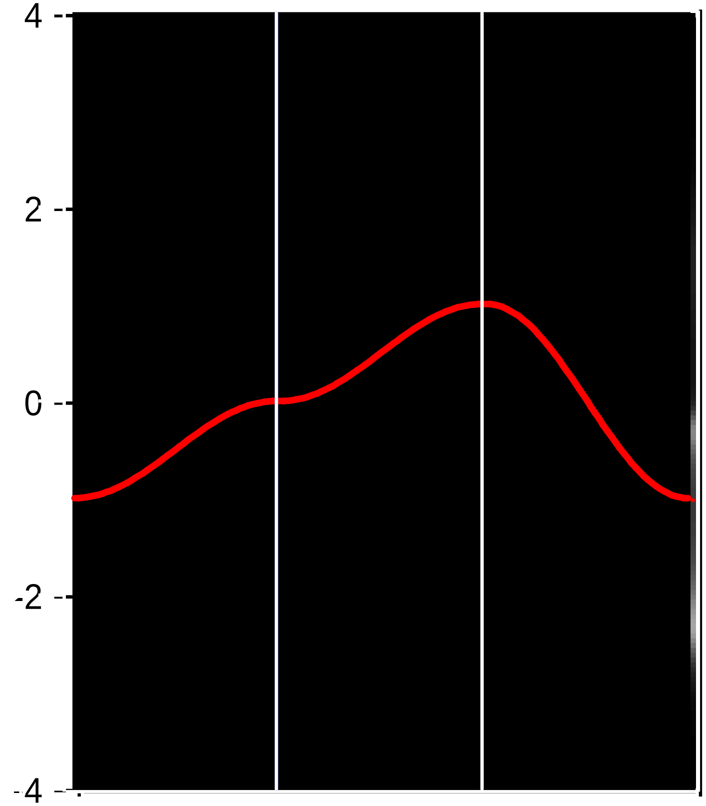
Self-consistent procedure to update SP energies – same effect as correcting them with Real Σ only (!)

METAL – INSULATOR TRANSITION IN MODEL SYSTEMS

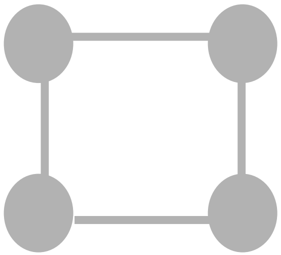
2D

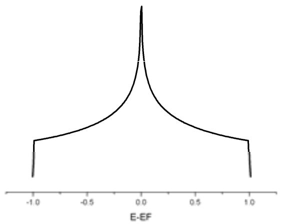




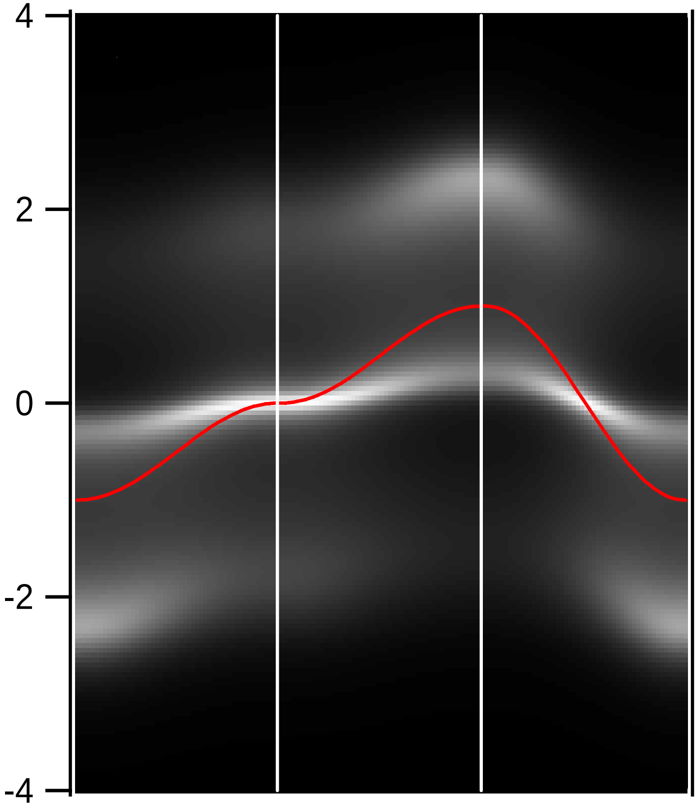


2D

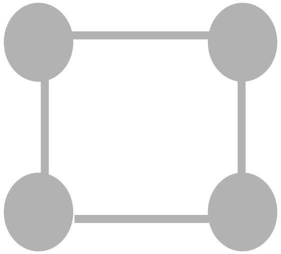


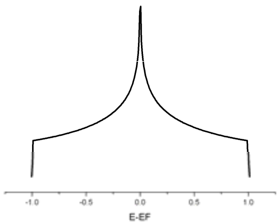


U=   W

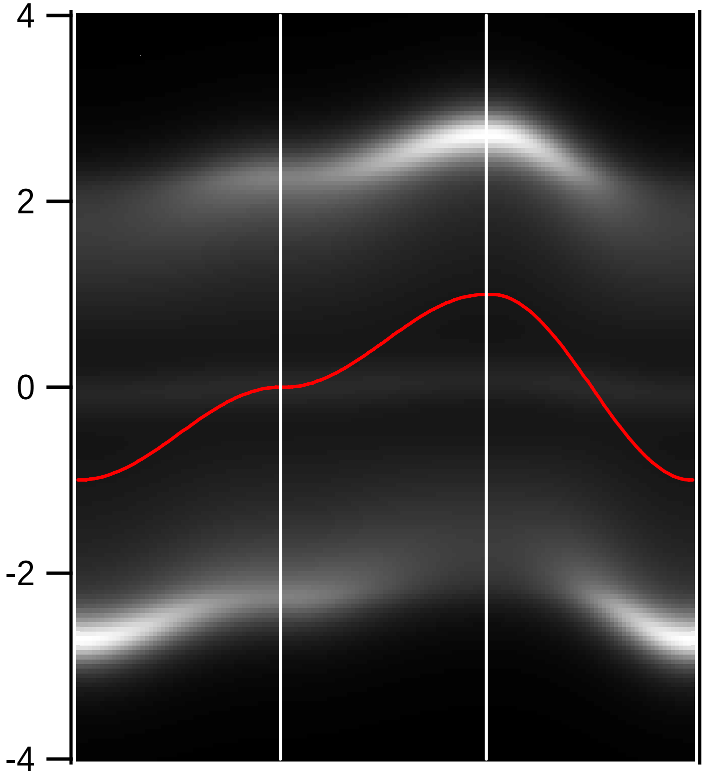


2D

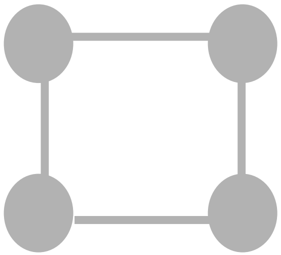


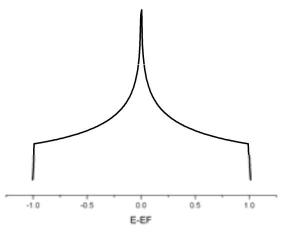


U=1.5W

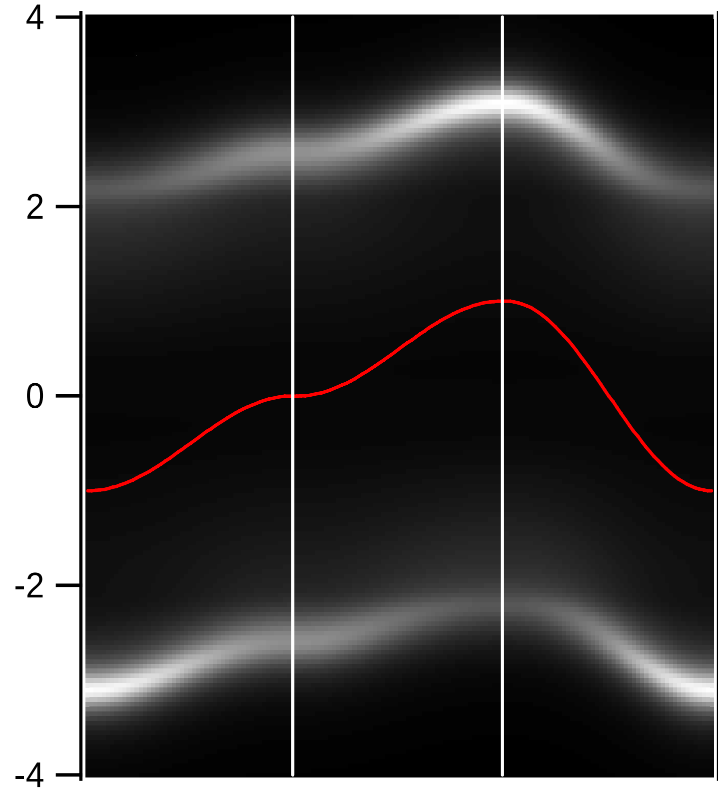


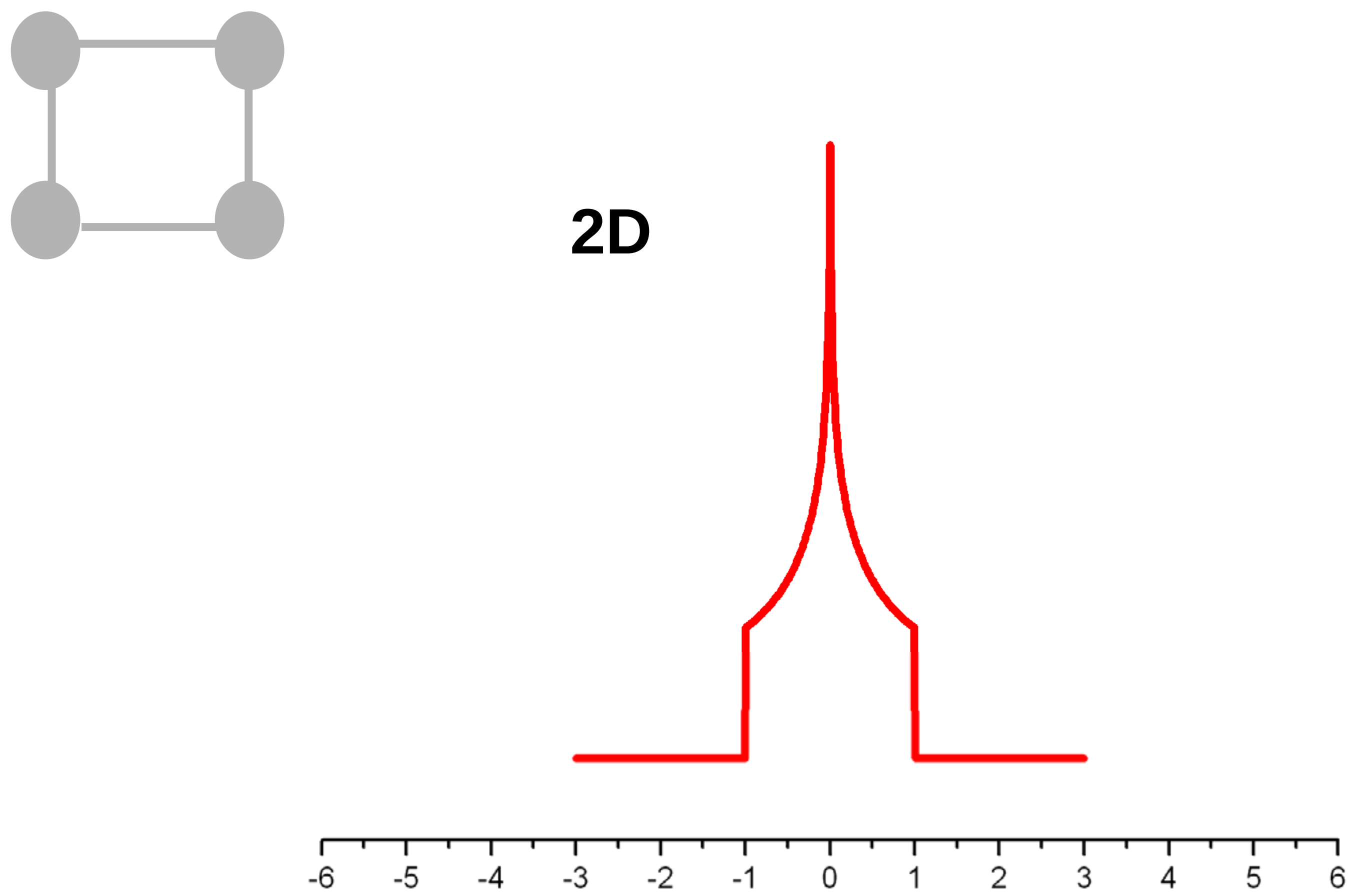
2D

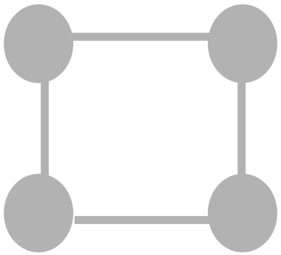


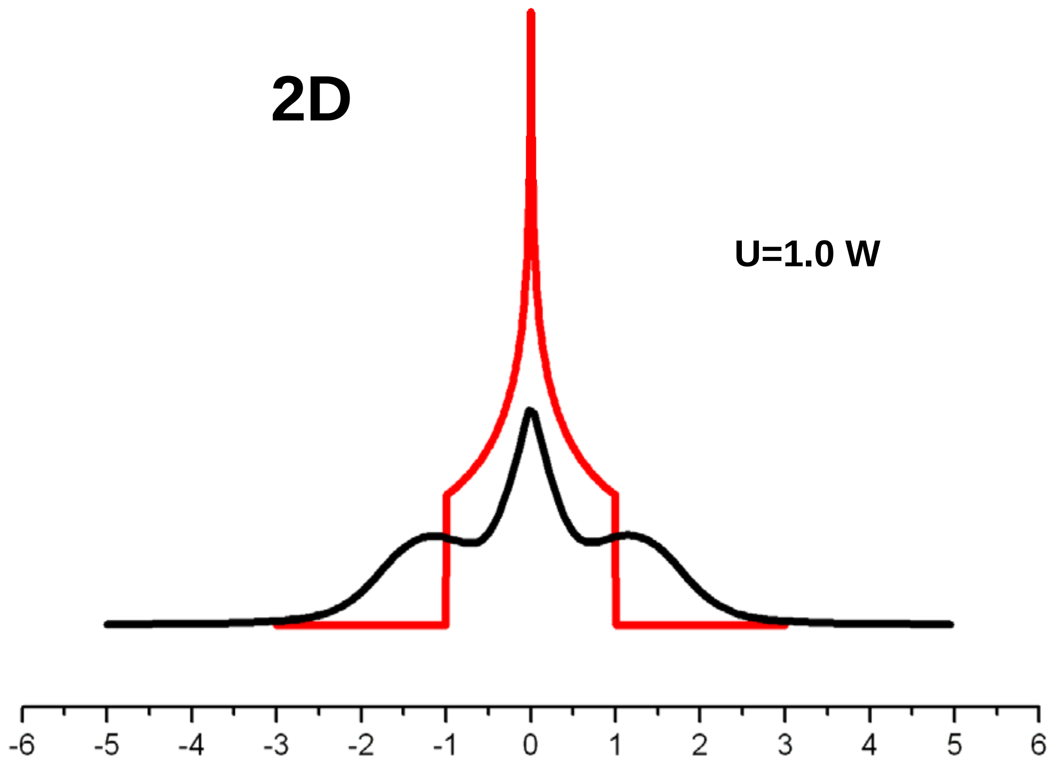


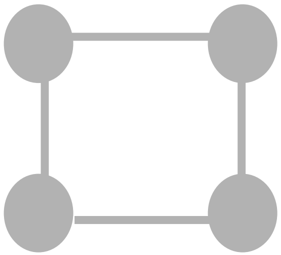
U=2 W

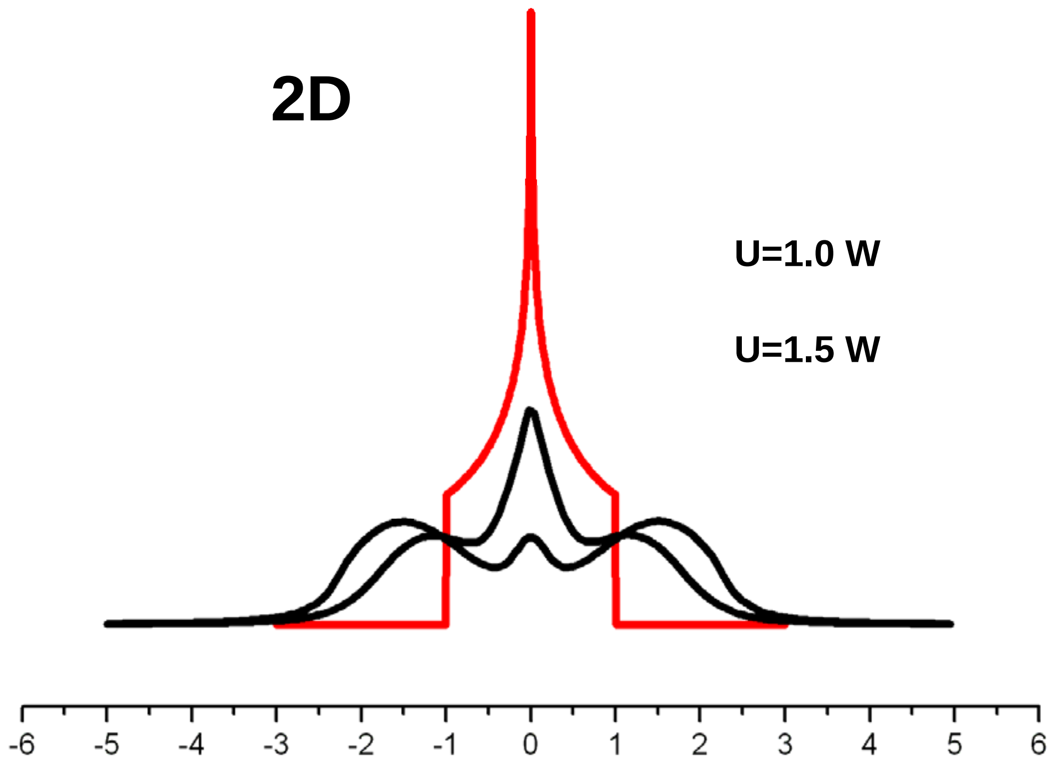


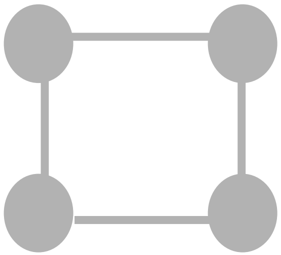

2D
-6 -5 -4 -3 -2 -1 0 1 2 3 4 5 6

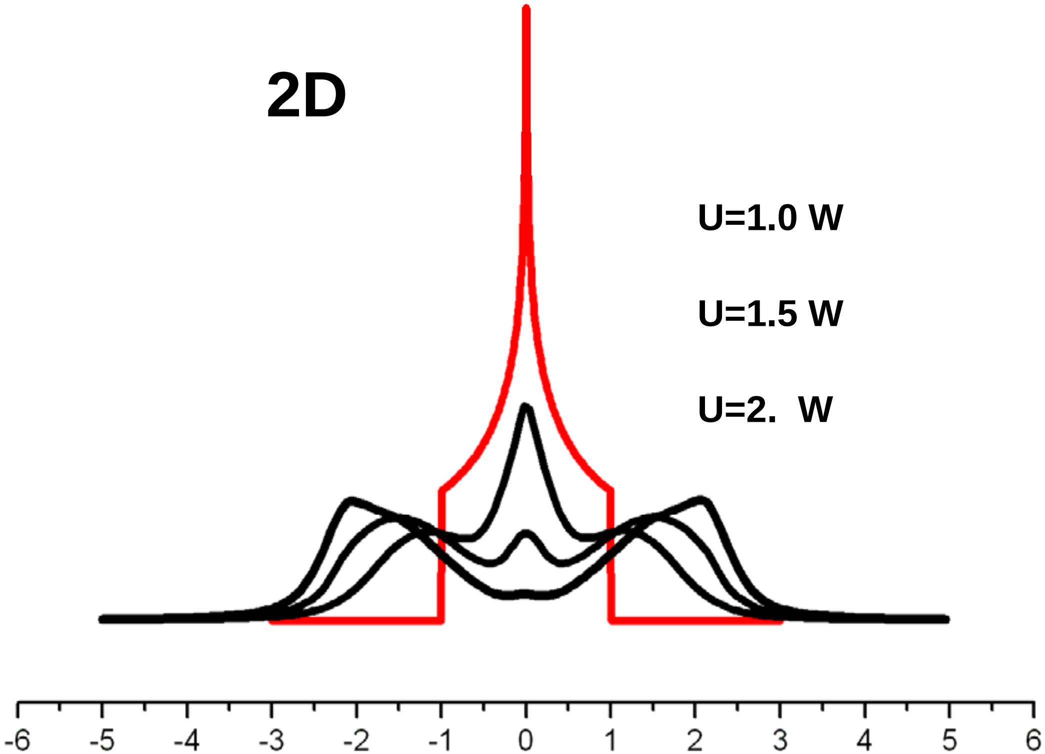



2D
U=1.0 W
-6
-5
-4
-3
-2
-1
0
1
2
3
4
5
6

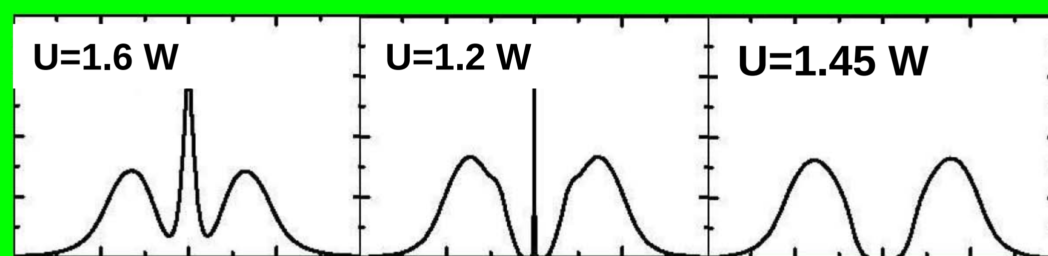



2D
U=1.0 W
U=1.5 W
-6
-5
-4
-3
-2
-1
0
1
2
3
4
5
6





DMFT results



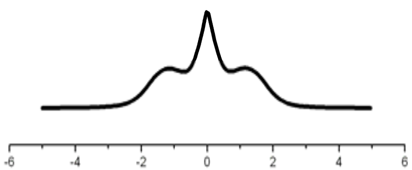
R.Bulla, PRL **83** 136 (1999):

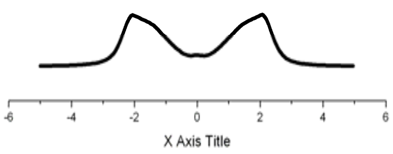
Metal Insulator Transition

U=W

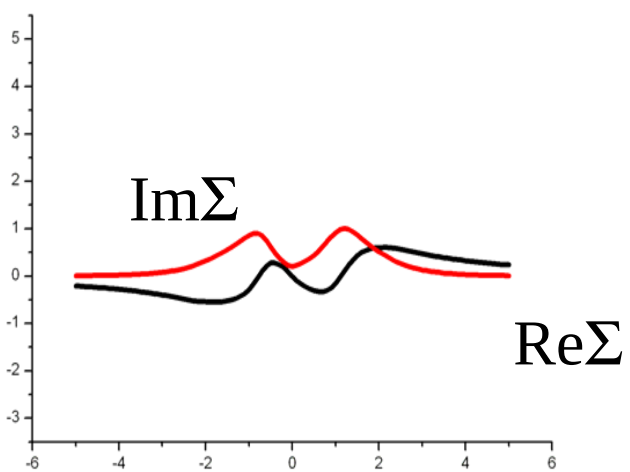
U=2W

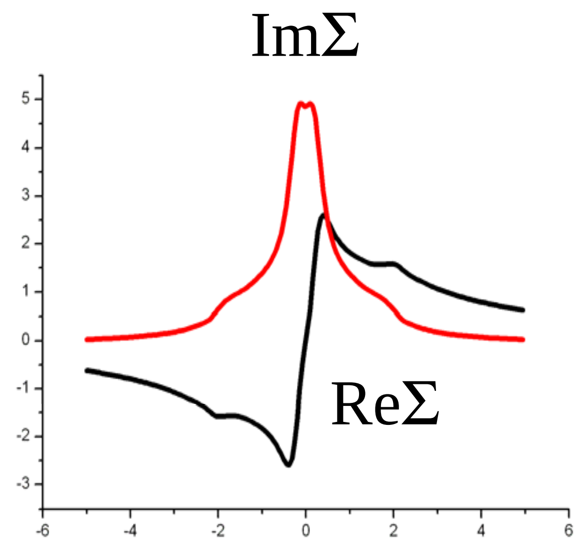
$$A(k,\omega) = \frac{1}{\pi} Im \frac{1}{\omega - \epsilon_0 - \Sigma(\omega)}$$





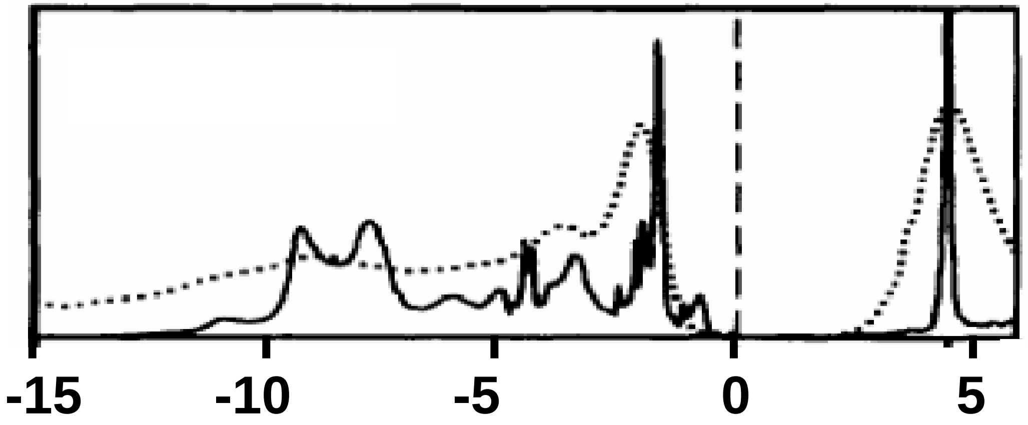
$\Sigma(\omega)$





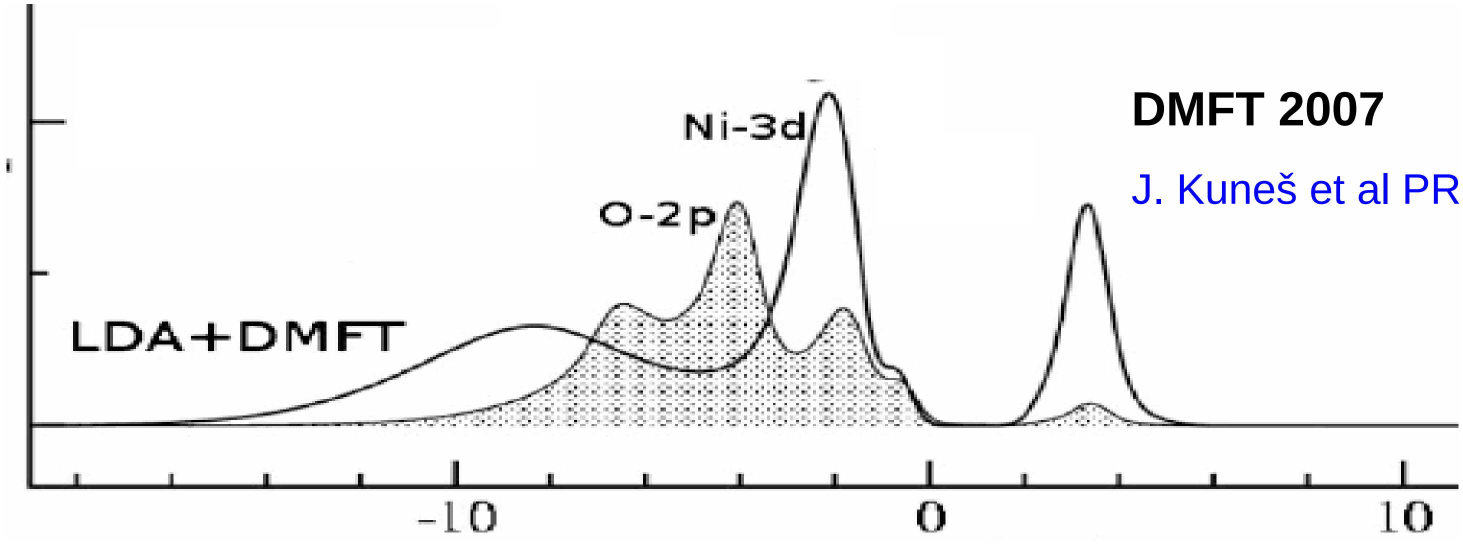
Real materials : DFT+3BS

Paramagnetic NiO



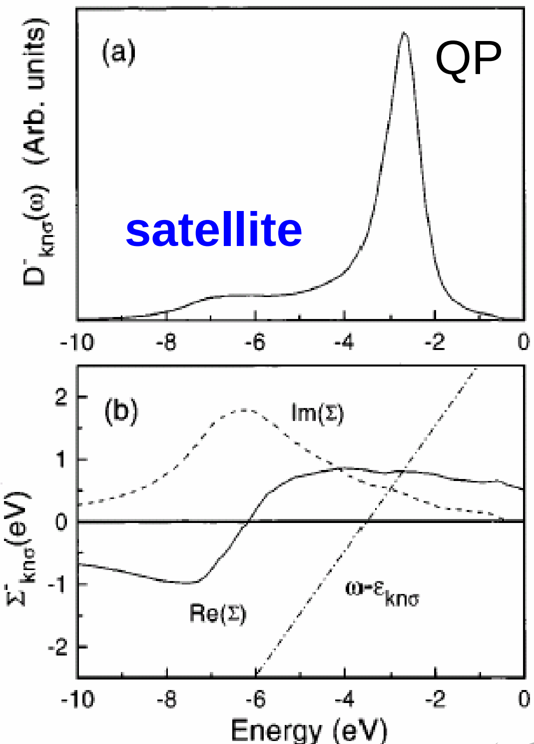
3BS 1994

FM et al PRL. **73**, 3129



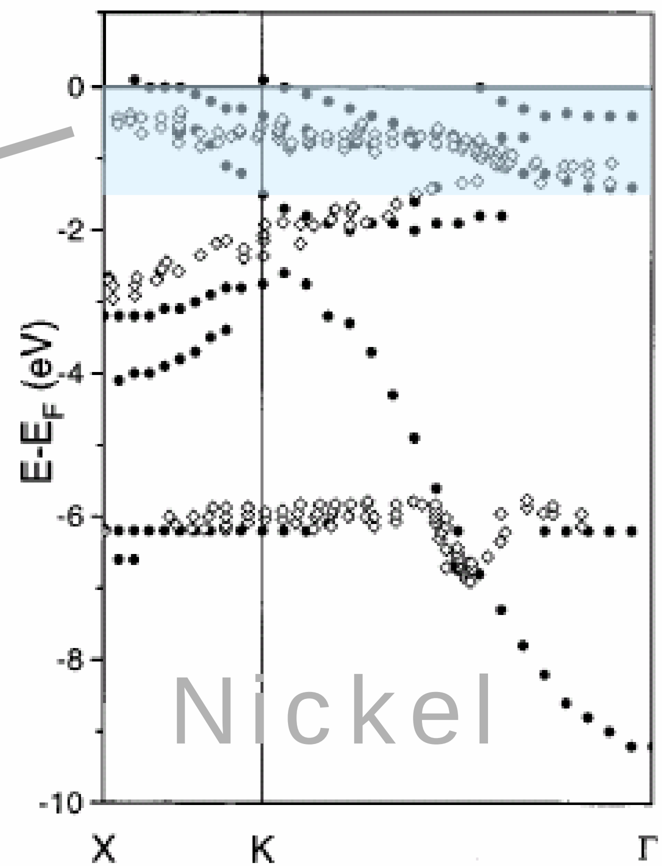
DMFT 2007

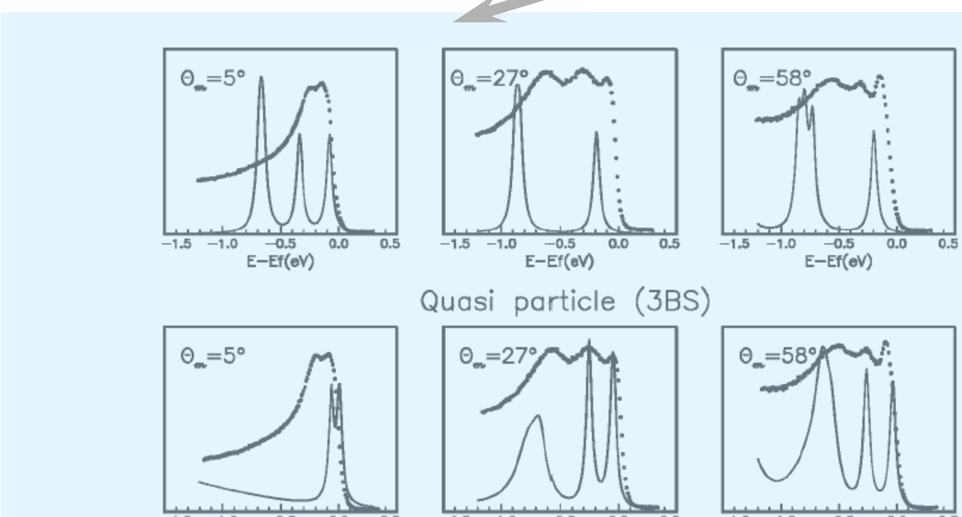
J. Kuneš et al PRL 99, 156404



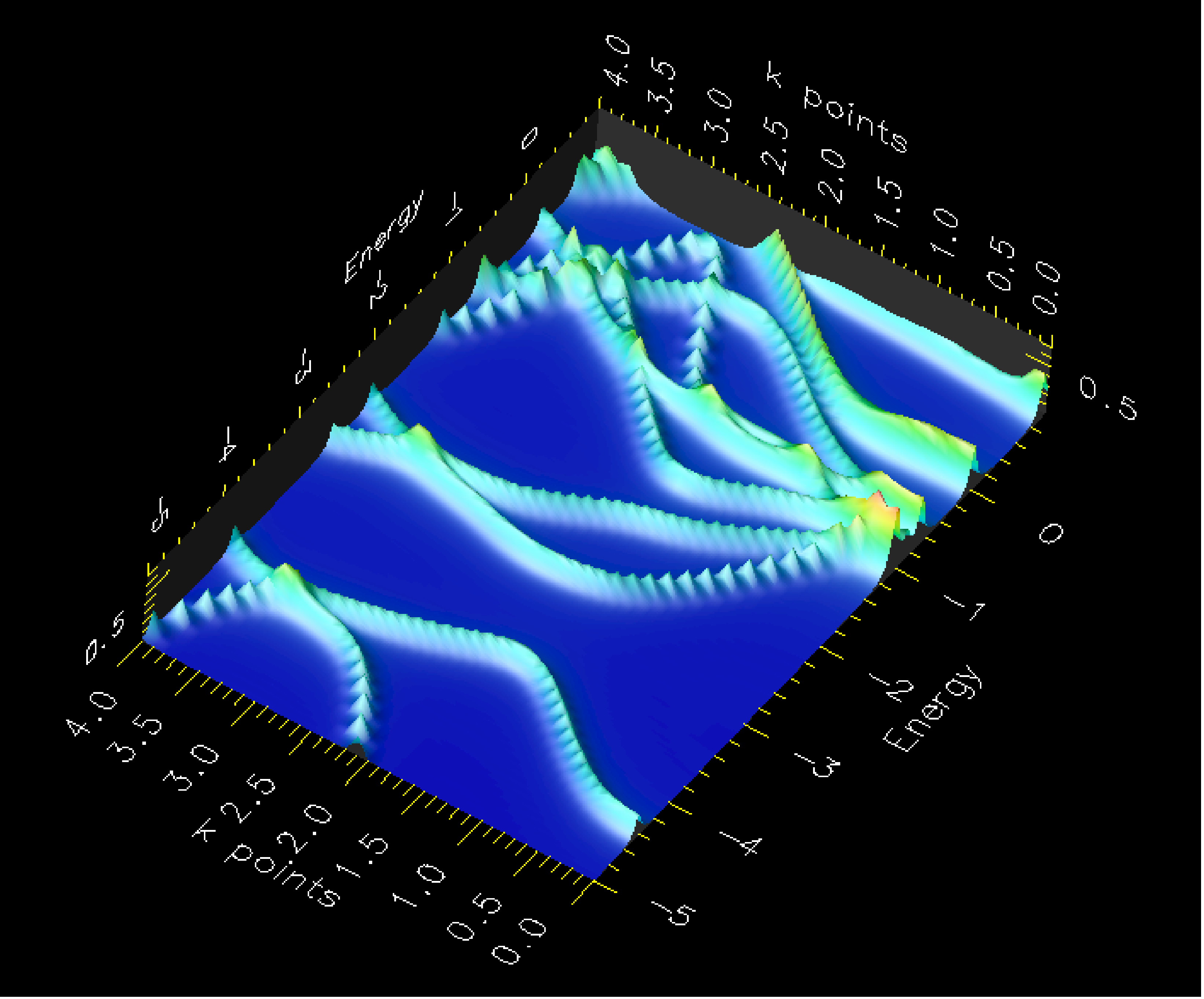
$$A(k,\omega) = \frac{1}{\pi} Im \frac{1}{\omega - \epsilon_{k0} - \Sigma(k\omega)}$$

QP band structure

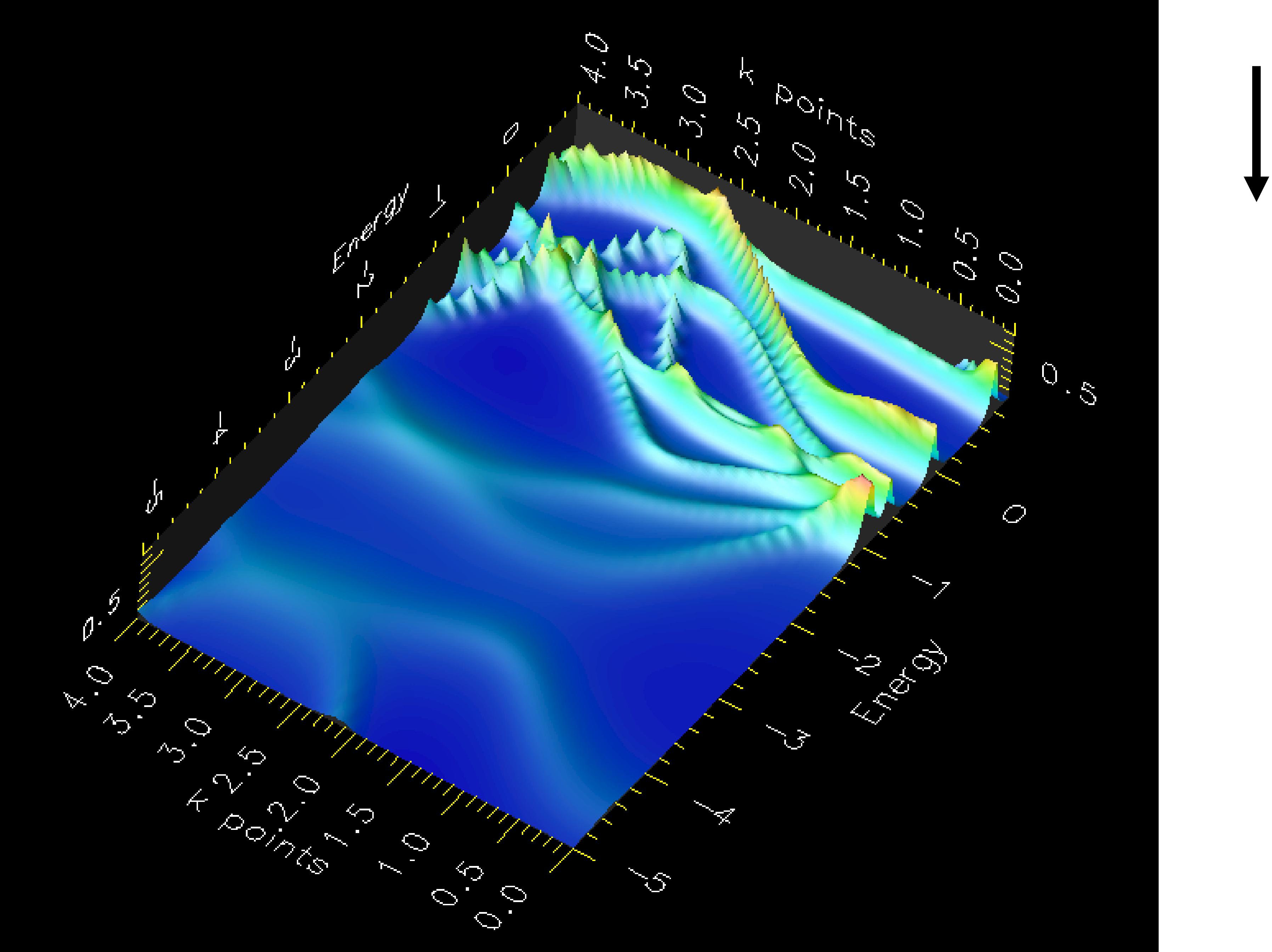




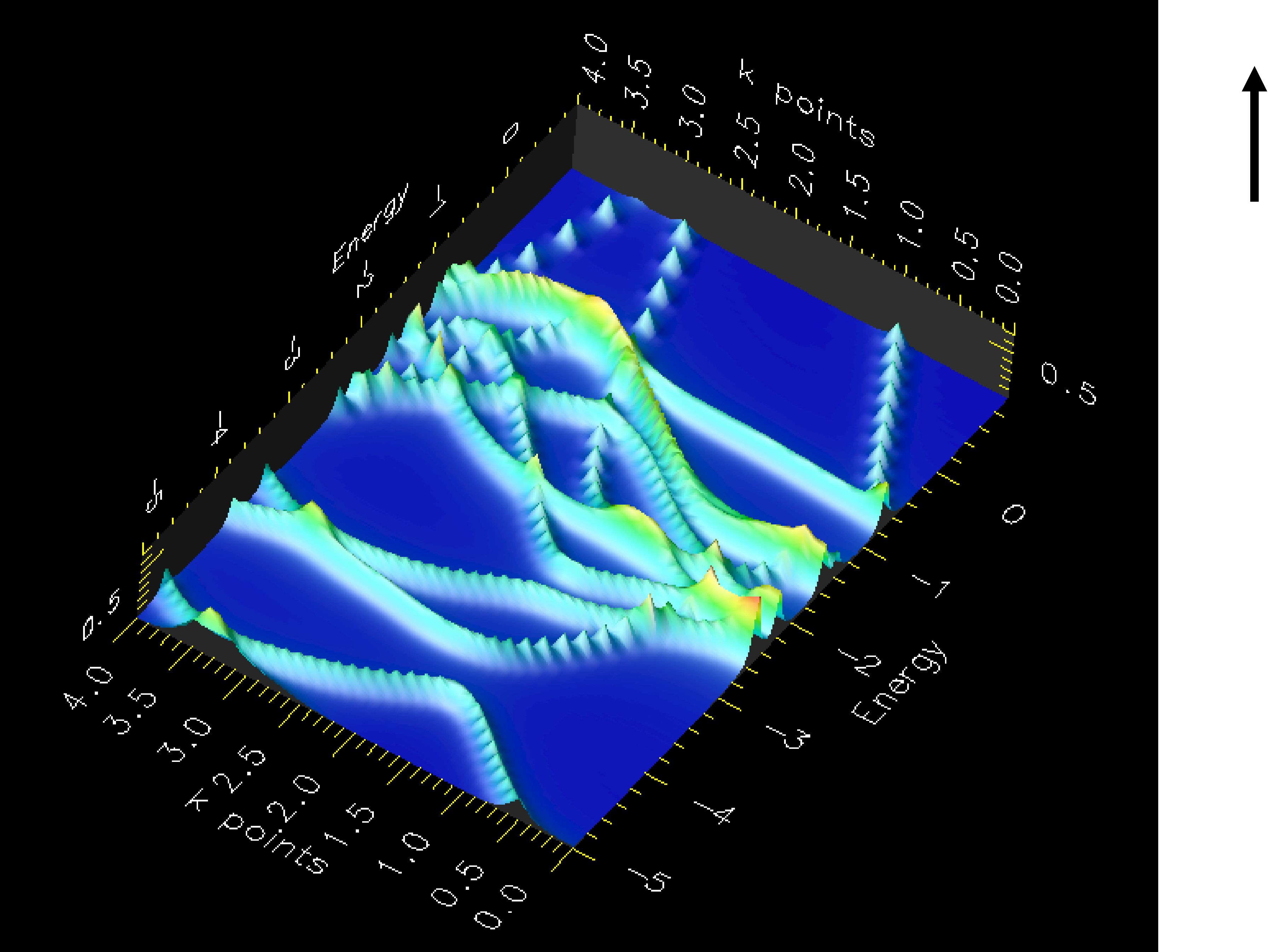
FM, V. Bellini, J. Osterwalder, T.J. Kreutz, P. Aebi, C. Arcangeli PRB 59, R10409 (1999)



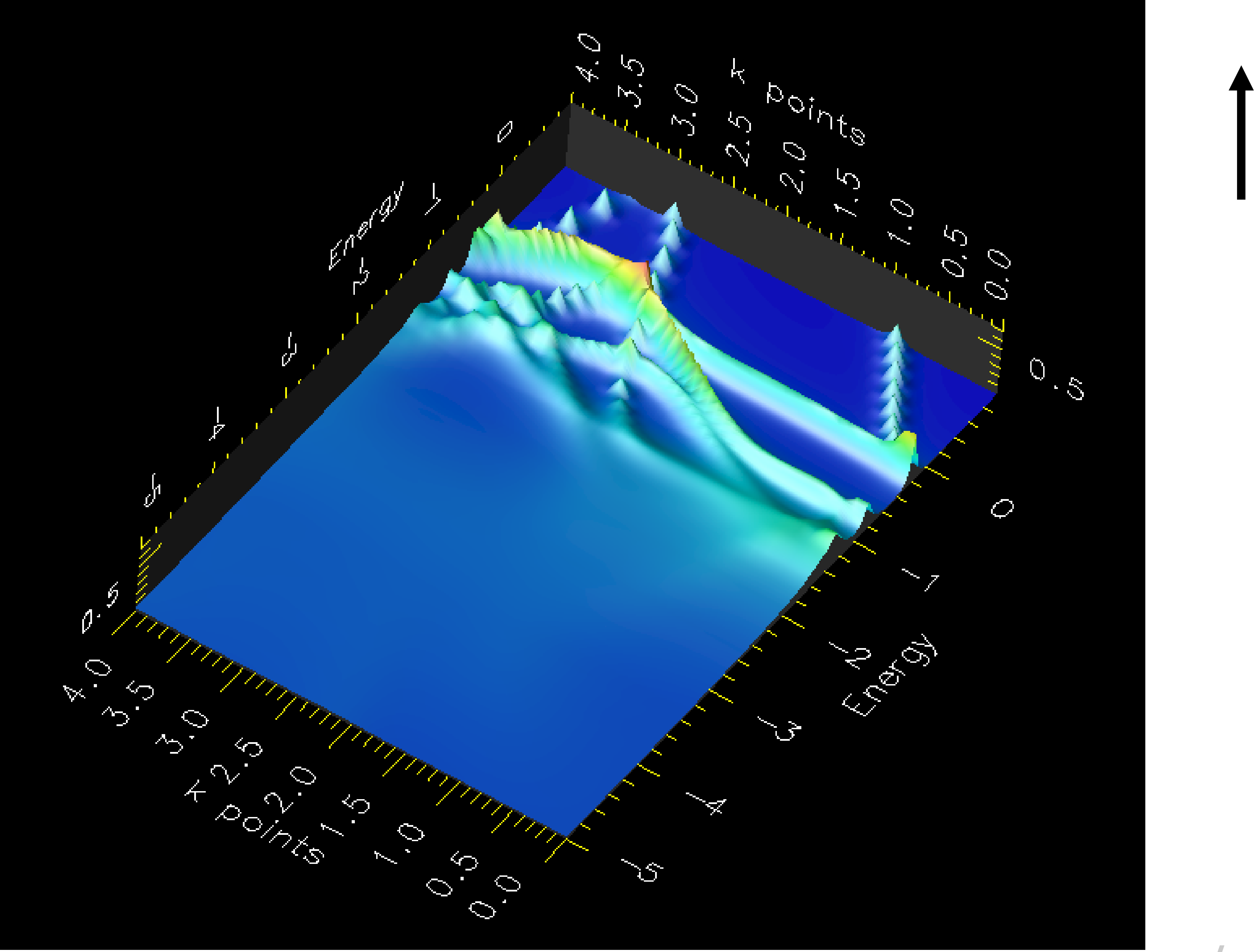
cobalt



cobalt



cobalt



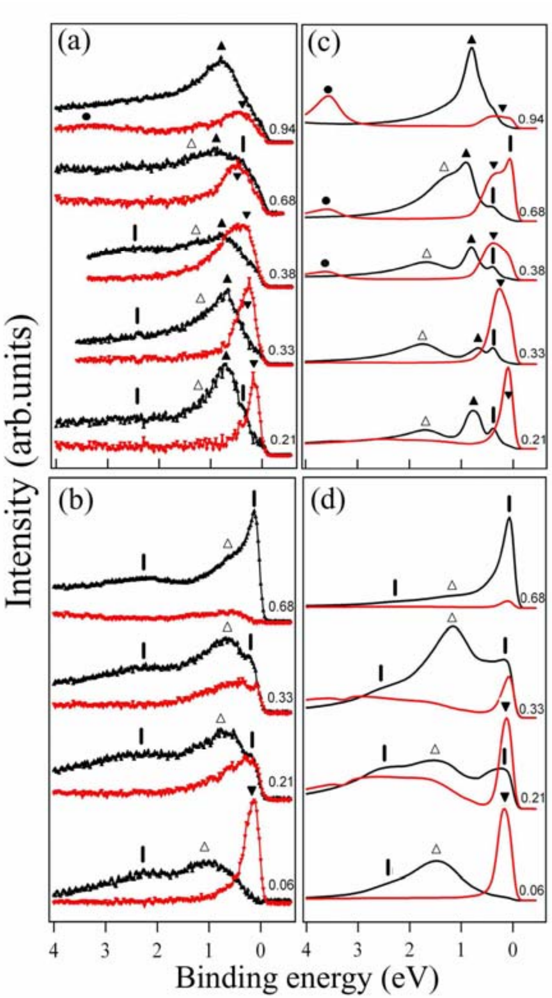
FM et a J. ELEC. SPECTROSCOPY REL. PHEN 137 523 2004

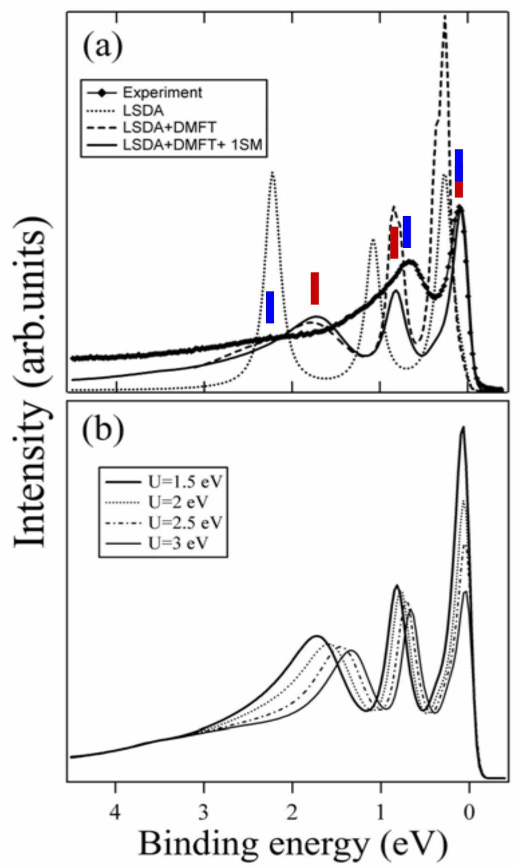
cobalt

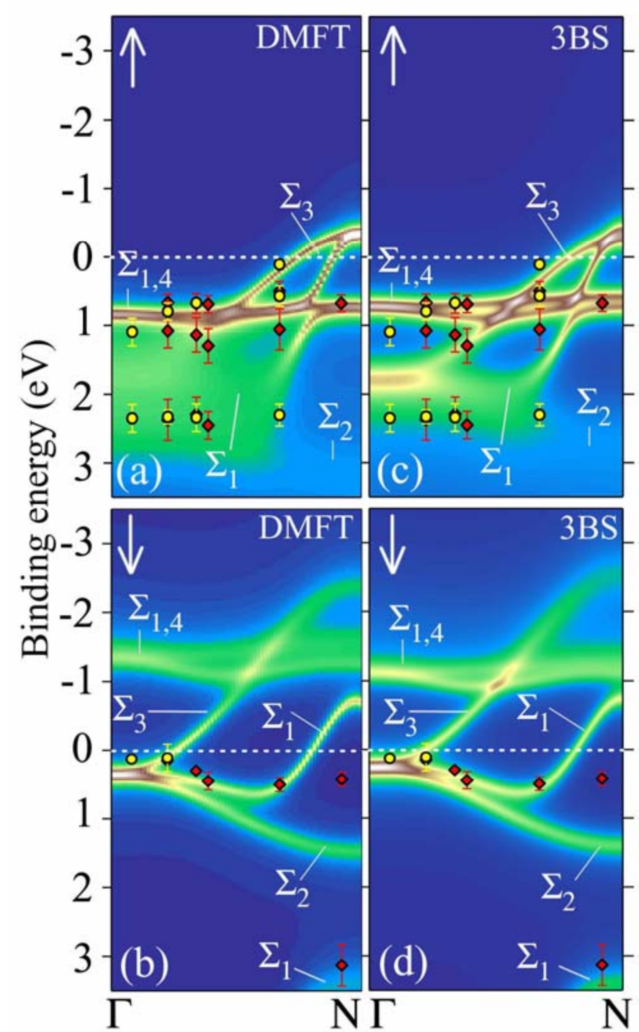
About the strength of correlation effects in the electronic structure of iron

J. Sánchez-Barriga[1], J. Fink[1,2], V. Boni [3], I. Di Marco[4,5], J. Braun[6], J. Minár[6], A. Varykhalov[1], O. Rader[1], V. Bellini[3], F. Manghi[3], H. Ebert[6], M.I. Katsnelson[5], A. I. Lichtenstein[7], O. Eriksson[4], W. Eberhardt[1], and H. A. Dürr[1]

exp theo







arXiv:0910.4360v1

Photoemission from core states:

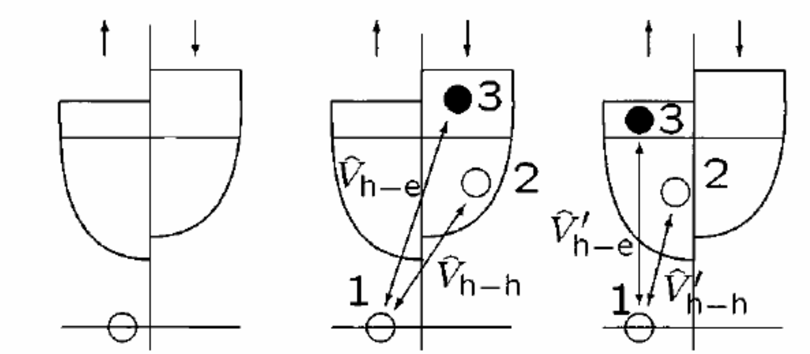
how to recover the core multiplet structure in the solid state

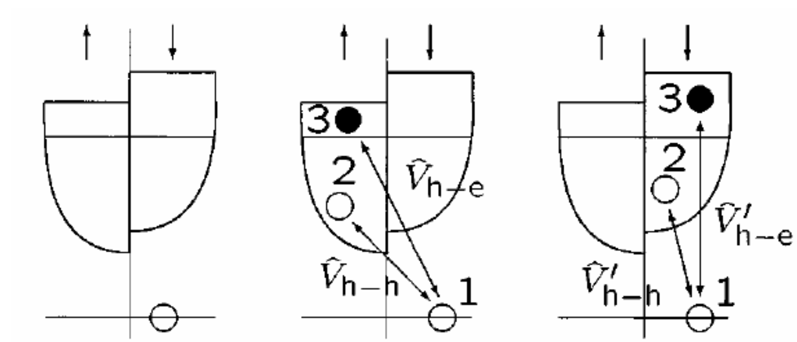
$$\hat{H} = \hat{H}^{vv} + \hat{H}^{cc} + \hat{H}^{cv}$$

$$\hat{H}^{vv} = \sum_{\mathbf{k}v\sigma} \epsilon^{v}_{\sigma}(\mathbf{k})\hat{n}^{v}_{\mathbf{k}\sigma}$$

$$\hat{H}^{cc} = \sum_{i\sigma} \epsilon^{c}_{\sigma}\hat{n}^{c}_{i\sigma} + \sum_{i\sigma} U^{cc}_{\sigma-\sigma}\hat{n}^{c}_{i\sigma}\hat{n}^{c}_{i-\sigma}$$

$$\hat{H}^{cv} = \sum_{iv\sigma} [U^{cv}_{\sigma-\sigma}\hat{n}^{c}_{i\sigma}\hat{n}^{v}_{i-\sigma} + (U^{cv}_{\sigma\sigma} - J^{cv}_{\sigma\sigma})\hat{n}^{c}_{i\sigma}\hat{n}^{v}_{i\sigma}]$$





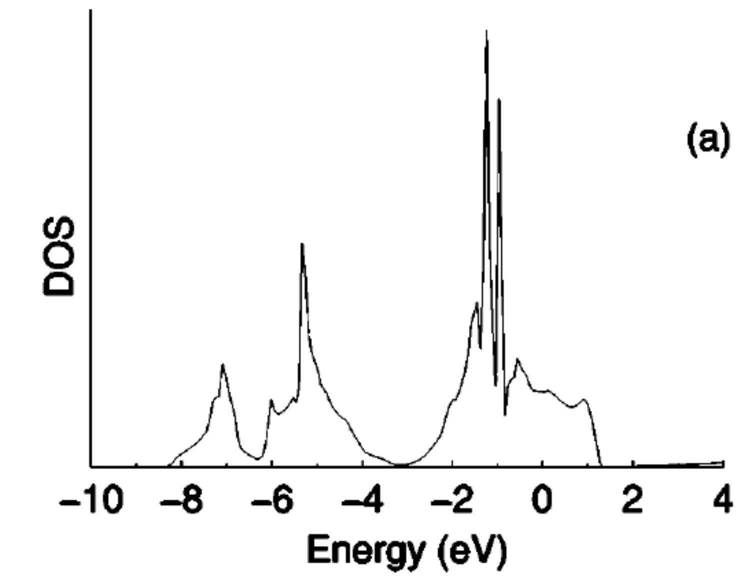
$$G_{c\sigma}(\omega) = -\frac{1}{\omega - \epsilon_{c\sigma}^{MF} - \Sigma_c(\omega)}$$

$$\epsilon_{c\sigma}^{\mathrm{MF}} = \epsilon_{c\sigma}^{H} + [(U_{cv} - J_{cv})\langle \hat{n}_{v\sigma} \rangle + U_{cv}\langle \hat{n}_{v-\sigma} \rangle] + U_{cc}\langle \hat{n}_{c-\sigma} \rangle$$

$$\boxed{\begin{aligned} \Sigma_c(\omega) = {} & \sum_d U_{cd}\frac{N_v^h}{N} - \sum_d \int_{E_F}^{+\infty} n_d(\epsilon) T_{hh}^{cd}(\omega - \epsilon) \\ & \times (1 + U_{cd}A_{cd})(\omega - \epsilon)\, d\epsilon + \sum_d (U_{cd} - J_{cd})\frac{N_v^h}{N} \\ & - \sum_d \int_{E_F}^{+\infty} n_d(\epsilon) T'^{cd}_{hh}(\omega - \epsilon) \\ & \times [1 + (U_{cd} - J_{cd})] A_{cd}(\omega - \epsilon)\, d\epsilon. \end{aligned}}$$

Core Level photoemission from Ni 2p in NiO

Rozzi, FM, Arcangeli, PRB 62 R4774 (2000)



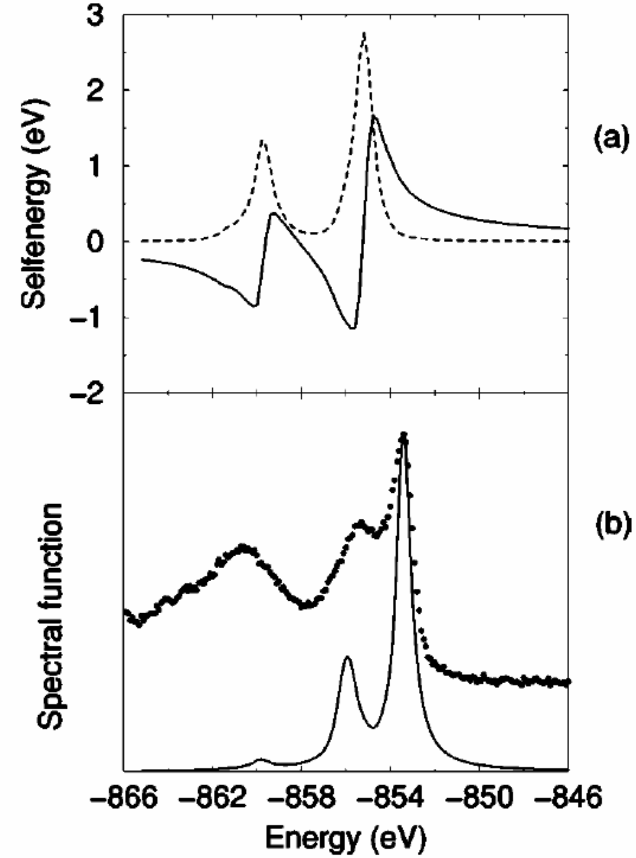
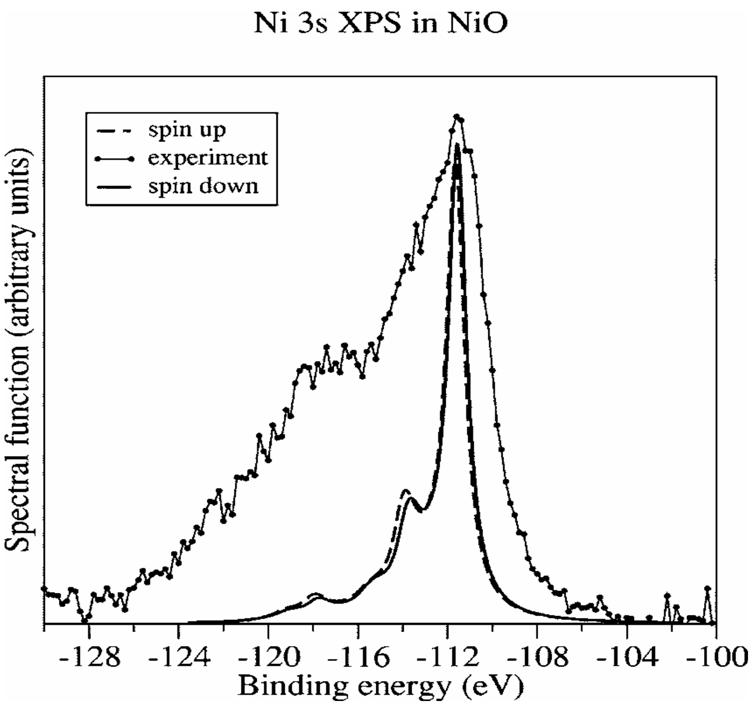


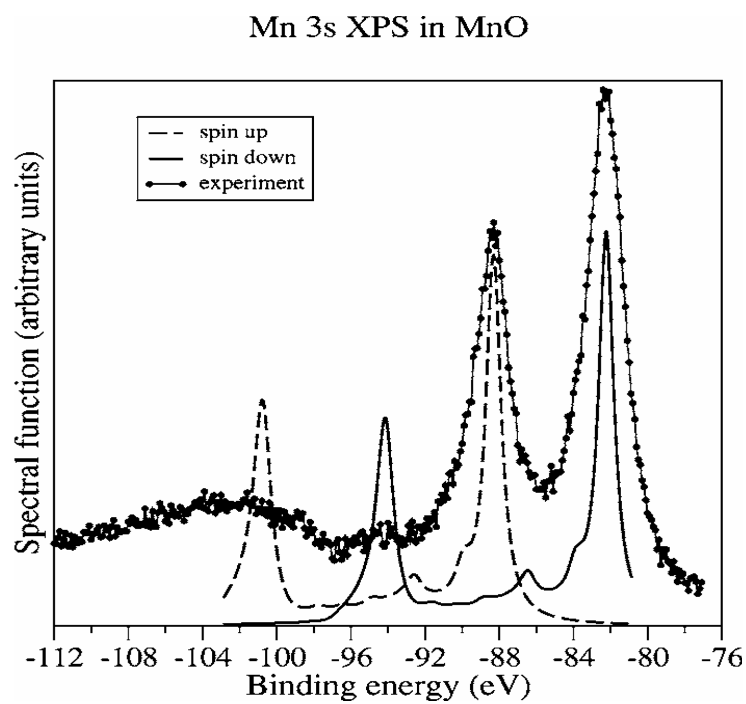
TABLE I. Coulomb integrals (eV) involving $2p$ and $3d$ Ni orbitals.

U_{cd}	d_{xy}	d_{yz}	d_{zx}	$d_{x^2-y^2}$	d_{z^2}
p_x	1.42	1.37	1.42	1.42	1.39
p_y	1.42	1.42	1.37	1.42	1.39
p_z	1.37	1.42	1.42	1.37	1.43

TABLE II. Exchange integrals (eV) involving $2p$ and $3d$ Ni orbitals.

J_{cd}	d_{xy}	d_{yz}	d_{zx}	$d_{x^2-y^2}$	d_{z^2}
p_x	0.05	0.01	0.05	0.05	0.02
p_y	0.05	0.05	0.01	0.05	0.02
p_z	0.01	0.05	0.05	0.01	0.06





3s line shapes can be reproduced within a solid-state picture that includes both the itinerant character of valence electrons and the atomic-like Coulomb interaction between valence and core states. Multiplet splitting -dominant in the case of MnO - is reproduced and interpreted as the result of a band-structure effect.

C. A. Rozzi, FM, and C. Calandra PRB 72, 125106 2005

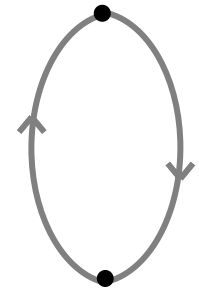
XAS and XMCD

Absorption cross section

$$\mu^{\pm}(\omega) \propto \sum_{kn} |D^{\pm}_{ckn}|^2 \sum_{\sigma} \chi_{ckn\sigma}(\omega)$$

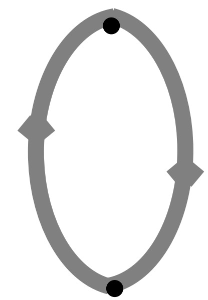
0. *Fermi golden rule*

$$\boxed{Im\chi^{00}_{ckn\sigma}(\omega) \propto \delta(\omega - (\epsilon_{kn\sigma} - \epsilon_{core}))}$$



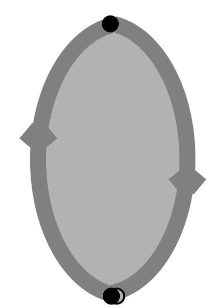
1. *Dress hole and particle propagators*

$$\boxed{Im\chi^{0}_{ckn\sigma}(\omega) \propto \int A^c_{\sigma}(\Omega) A^v_{kn\sigma}(\Omega + \omega) d\Omega}$$

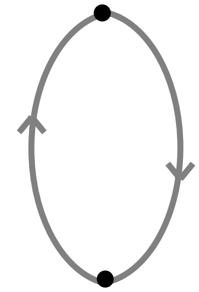


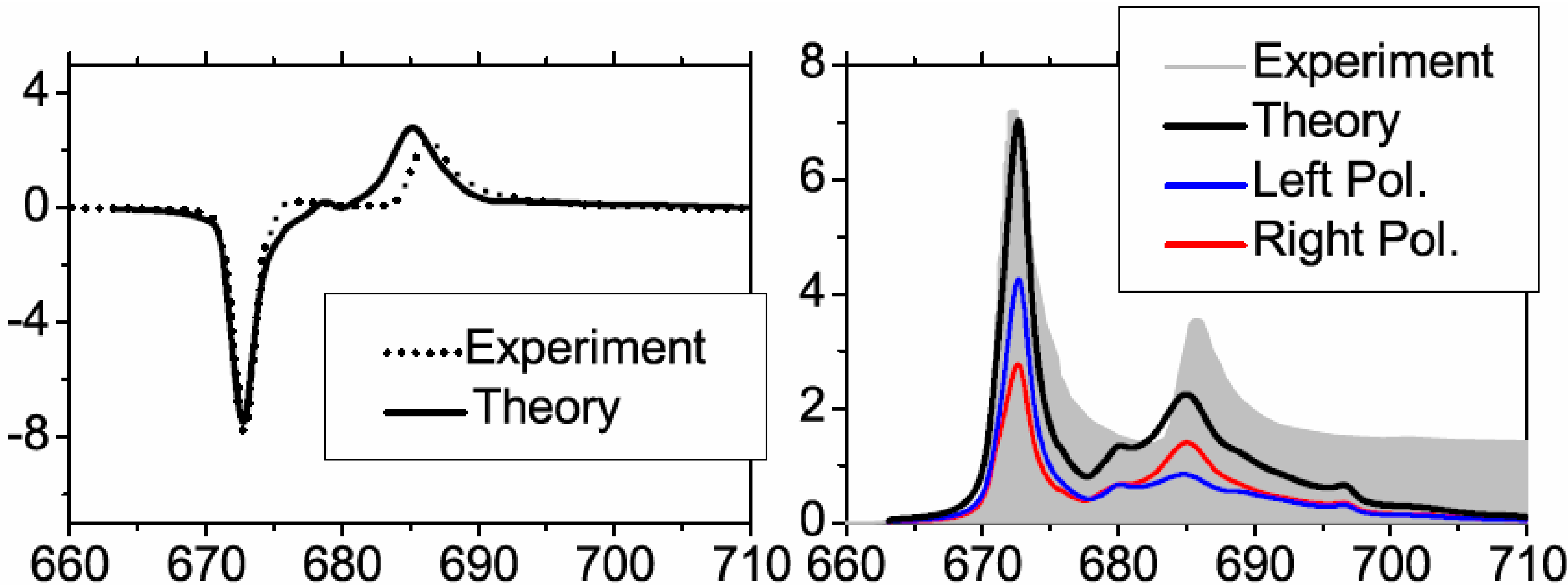
2. *Let hole and particle interact*

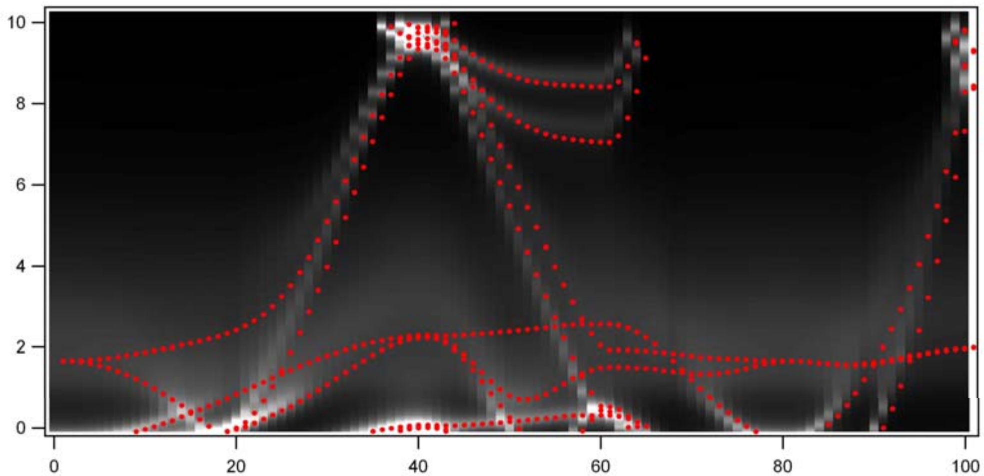
$$\boxed{\chi_{ckn\sigma}(\omega) = \frac{\chi^0_{ckn\sigma}(\omega)}{1 - \chi^0_{ckn\sigma}(\omega) T_{kn\sigma}(\omega)}}$$

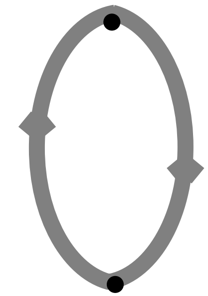


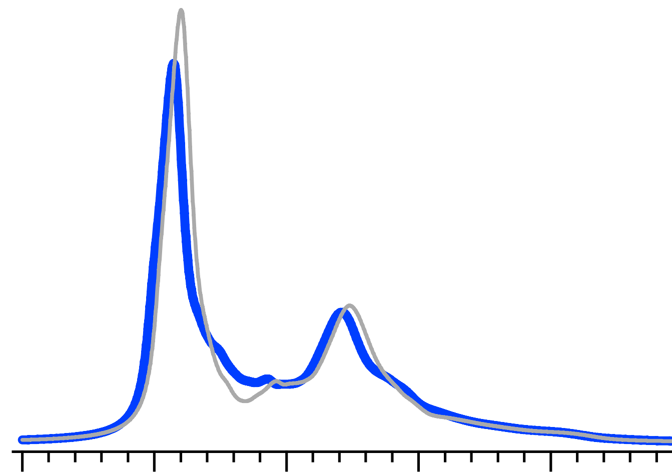
XMCD at the $L_{2,3}$ edge of iron

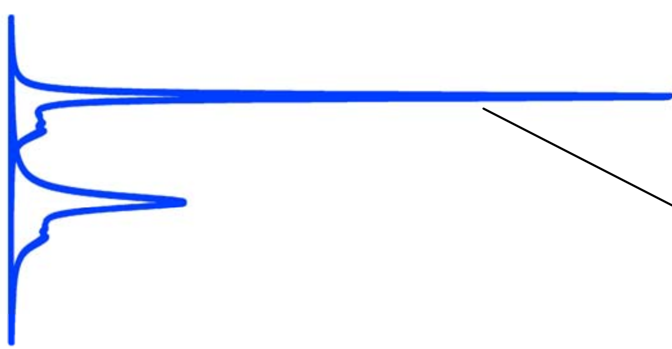












$$Im\chi^0_{ckn\sigma}(\omega) \propto \int A^c_\sigma(\Omega) A^v_{kn\sigma}(\Omega+\omega)\, d\Omega$$

h-e attraction

assume the excited states of the N-particle interacting system to be a superposition of single particle states with one core hole and an electron (Tamm-Dancoff) and express the Hamiltonian in this basis to get two particle eigenvalue in the presence of e-e interaction

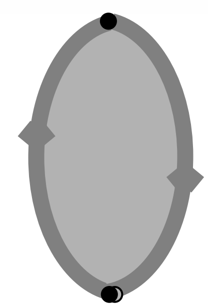
$$E_{ckn\sigma} = E^0_{ckn\sigma} + T_{kn\sigma}(\omega) \quad \textit{where} \quad T_{kn\sigma}(\omega) = \sum_{\alpha} \frac{-U_{pd}|C^n_{\alpha\sigma}(k)|^2}{1 - U_{pd}\chi^0_{\alpha\sigma}(\omega)}$$

Since the two-particle energies enter the definition of the two-particle propagator as

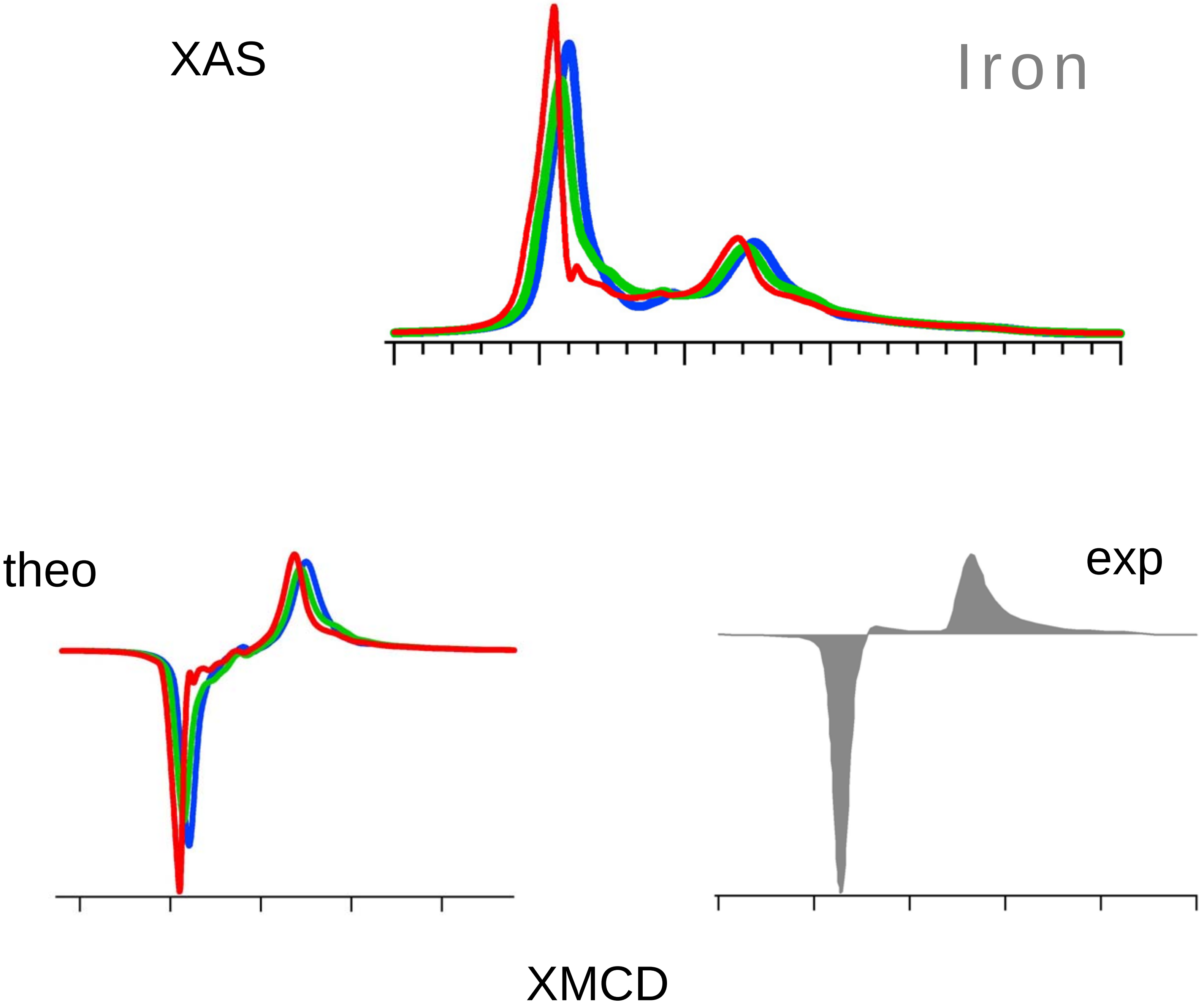
$$\chi_{ckn\sigma}(\omega) = \frac{-\imath}{\omega - E_{ckn\sigma} + \imath\eta} \qquad \chi^0_{ckn\sigma}(\omega) = \frac{-\imath}{\omega - E^0_{ckn\sigma} + \imath\eta}$$

we get

$$\boxed{\chi_{ckn\sigma}(\omega) = \frac{\chi^0_{ckn\sigma}(\omega)}{1 - \chi^0_{ckn\sigma}(\omega)T_{kn\sigma}(\omega)}}$$



Bethe Salpeter eqn. $$Q_{ckn\sigma}(\omega) = Q^0_{ckn\sigma}(\omega) + Q^0_{ckn\sigma}(\omega)T_{kn\sigma}(\omega)Q_{ckn\sigma}(\omega)$$


XAS
Iron
theo
exp
XMCD

Collaborators:

Valerio Bellini

Carlo Andrea Rozzi — *3BS development*

Andrea Ferretti

Valentina Boni

Lorenzo Pardini — *XAS and XMCD*

Claudia Ambrosch-Draxl

QUANTUM SOLIDS

http://www.quantumsolids.unimore.it

Summary

- *3BS is an efficient way to describe short range e-e correlations in model systems and real materials in all correlation regimes*
- *It describes quasi particle energy renormalization (real Σ) as well as incoherent states, QP quenching, MI transition (Im Σ)*
- *Results for core and valence states (hole and particle propagators)*
- *Can be extended to describe neutral excitaions (XAS and XMCD) and to include excitonic effects*