Angle-resolved photoemission from ferromagnetic 3d-systems: A combination of the one-step model with the self-consistent LSDA + DMFT approach

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Acknowledgement

Theory:

- J. Minár, S. Chadov, H. Ebert: Uni. München
- A. Lichtenstein: Uni. Hamburg
- M. Katsnelson: Uni. Nijmegen

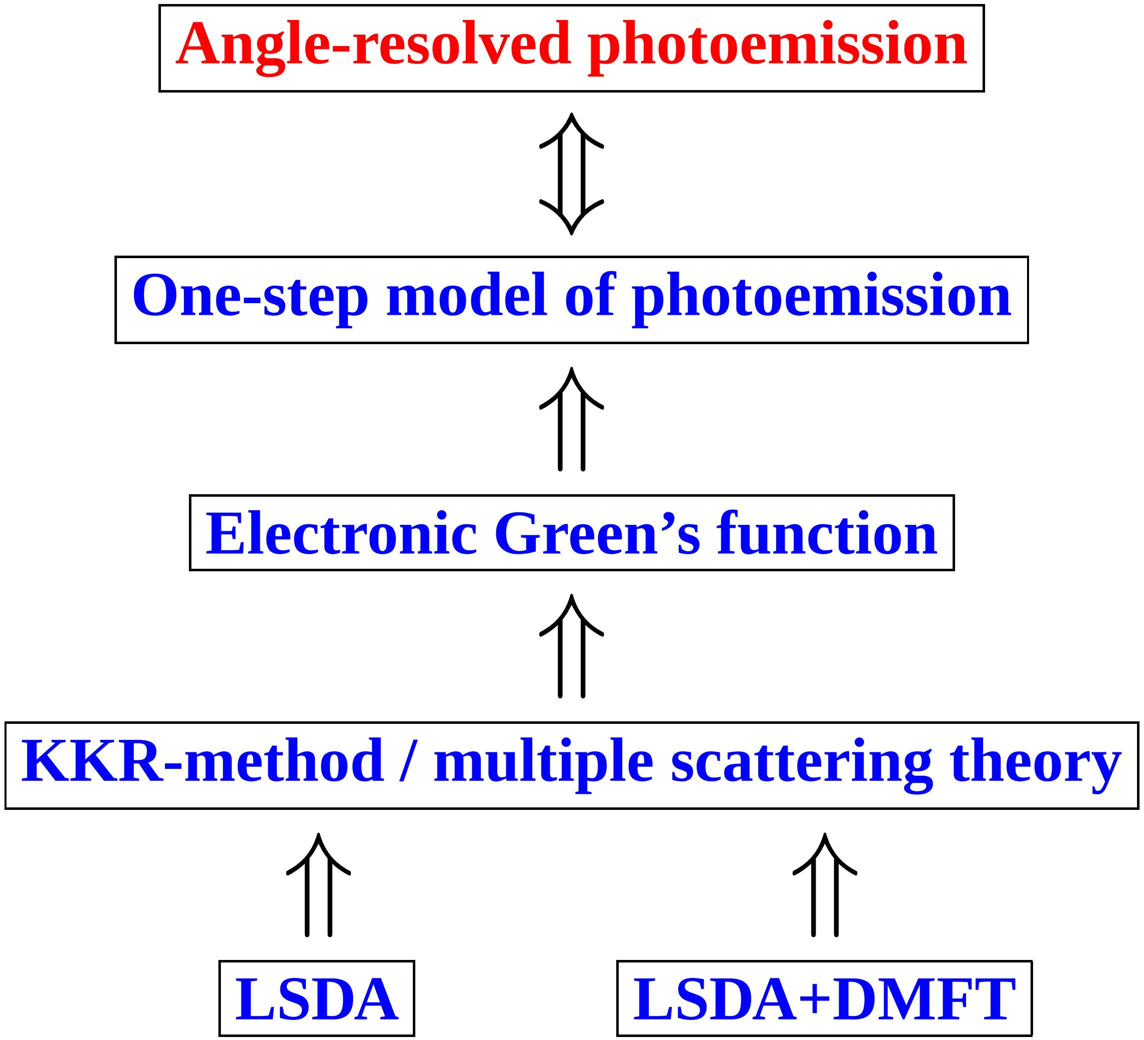
Experiments:

- Ch. Fadley, L. Plucinski: Uni. California Davis
- M. Mulazzi, G. Panaccione: Elettra, Trieste

Outline

- Theoretical aspects of spinpolarized photoemission
- Ni(011): Electronic structure and angle-resolved UV-photoemission
- Fe(001): Electronic structure and angle-resolved UV-photoemission
- Ni(001): Fermi surface and angle-resolved X-ray photoemission
- Conclusions and outlook

LSDA+DMFT and photoemission



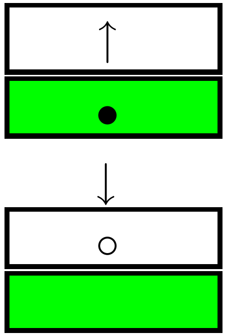
General theory of photoemission

Fermi's Golden Rule

$$\Gamma \;=\; -\;\frac{2\pi}{\hbar}\,|<\Psi_F|\Delta|\Psi_I>|^2\,\delta(E_F - E_I - \epsilon_{ph})$$

$$\Delta^{PES} \;=\; \sum_{\mathbf{e},\mathbf{k}} M^P_{\mathbf{e},\mathbf{k}}\; a^\dagger_{\mathbf{e}} a_{\mathbf{k}}$$

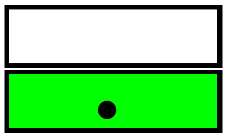
$$\Delta^{IPE} \;=\; \sum_{\mathbf{e},\mathbf{k}} M^P_{\mathbf{k},\mathbf{e}}\; a^\dagger_{\mathbf{k}} a_{\mathbf{e}}$$

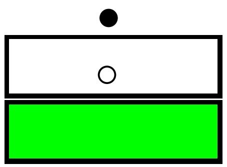


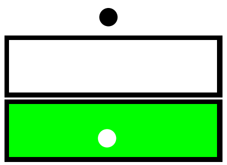
$$M^P_{\mathbf{e},\mathbf{k}} \;=\; <\phi^{SP}_{\mathbf{e}}|\mathbf{A_o}\cdot\mathbf{p}|\phi_{\mathbf{k}}>$$

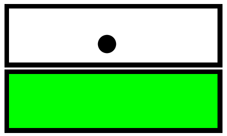
Sudden approximation

The interaction of the photoelectron with the rest system is neglected

PES:  $|\Psi_I> = |\Psi_0^N>$

IPE:  $|\Psi_I> = a^\dagger_{\mathbf{e}}|\Psi_0^N>$

PES:  $|\Psi_F> = a^\dagger_{\mathbf{e}}|\Psi_S^{N-1}>$

IPE:  $|\Psi_F> = |\Psi_S^{N+1}>$

One-step model

Inserting $|\Psi_I>$ and $|\Psi_F>$ in Fermi's Golden Rule
Summation over all possible final states
Averaging in the Grand Canonical Ensemble

$$\frac{1}{2\pi} < [T^\dagger(t), T(t')]_+ > \; = A^{(1)}(t,t') = \frac{1}{2\pi\hbar}\int dE e^{-\frac{i}{\hbar}E(t-t')}\, \mathrm{A}^{(1)}(\mathrm{E})$$

$$T^{PES} = \sum_{\mathbf{k}} M^P_{\mathbf{e},\mathbf{k}}\, a_{\mathbf{k}} \qquad T^{IPE} = \sum_{\mathbf{k}} M^P_{\mathbf{e},\mathbf{k}}\, a^\dagger_{\mathbf{k}}$$

One step model of photoemission

$$I(\epsilon_e, \mathbf{k}_\parallel) = \int d\mathbf{r} \int d\mathbf{r}'\; \Psi_e^\dagger(\mathbf{r})\; \boldsymbol{\alpha}\mathbf{A}_0\; A^{(1)}(\mathbf{r},\mathbf{r}',E)\; (\boldsymbol{\alpha}\mathbf{A}_0)^\dagger\; \Psi_e(\mathbf{r}')$$

$\hat{\boldsymbol{\alpha}} \cdot \mathbf{A}_0$ **: relativistic form of electron-photon interaction**

Initial and final states

Calculation of the initial states for $\Sigma^{DMFT}(E) \neq 0$

Relativistic LDA-Hamiltonian

$$h_{\mathrm{LDA}}(\mathbf{r}) = -ic\boldsymbol{\alpha}\boldsymbol{\nabla} + \beta c^2 - c^2 + V_{\mathrm{LDA}}(\mathbf{r}) + \beta\boldsymbol{\sigma}\mathbf{B}_{\mathrm{LDA}}(\mathbf{r})$$

$$V_{\mathrm{LDA}}(\mathbf{r}) = \frac{1}{2}(V^{\uparrow}_{\mathrm{LDA}}(\mathbf{r}) + V^{\downarrow}_{\mathrm{LDA}}(\mathbf{r})) \qquad \mathbf{B}_{\mathrm{LDA}}(\mathbf{r}) = \frac{1}{2}(V^{\uparrow}_{\mathrm{LDA}}(\mathbf{r}) - V^{\downarrow}_{\mathrm{LDA}}(\mathbf{r}))\mathbf{b}$$

Generalized nonlocal potential

$$U(\mathbf{r},\mathbf{r}',E) = \delta(\mathbf{r}-\mathbf{r}')\,(V_{\mathrm{LDA}}(\mathbf{r}) \;+\; \beta\boldsymbol{\sigma}\mathbf{B}_{\mathrm{LDA}}(\mathbf{r})) + V(\mathbf{r},\mathbf{r}',E) \;+\; \beta\boldsymbol{\sigma}\mathbf{B}(\mathbf{r},\mathbf{r}',E)$$

$$V(\mathbf{r},\mathbf{r}',E) = \frac{1}{2}(\Sigma^{\uparrow}(\mathbf{r},\mathbf{r}',E) + \Sigma^{\downarrow}(\mathbf{r},\mathbf{r}',E)) \quad \mathbf{B}(\mathbf{r},\mathbf{r}',E) = \frac{1}{2}(\Sigma^{\uparrow}(\mathbf{r},\mathbf{r}',E) - \Sigma^{\downarrow}(\mathbf{r},\mathbf{r}',E))\,\mathbf{b}$$

Dyson equation for the initial state Green function

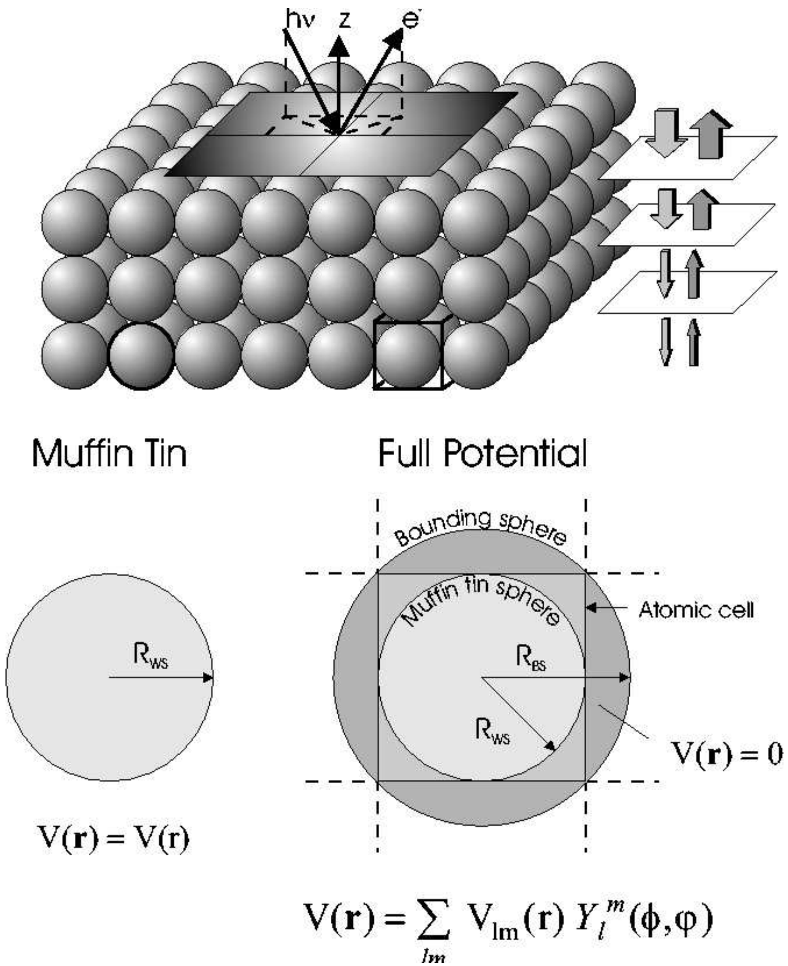
$$\left[E + \mu_0 + ic\boldsymbol{\alpha}\boldsymbol{\nabla} - \beta c^2 + c^2)\right] G_1^{+}(\mathbf{r},\mathbf{r}',E) + \int U(\mathbf{r},\mathbf{r}'',E)G_1^{+}(\mathbf{r}'',\mathbf{r}',E)d\mathbf{r}'' = \delta(\mathbf{r}-\mathbf{r}')$$

Time reversed SPLEED state for the photoelectron

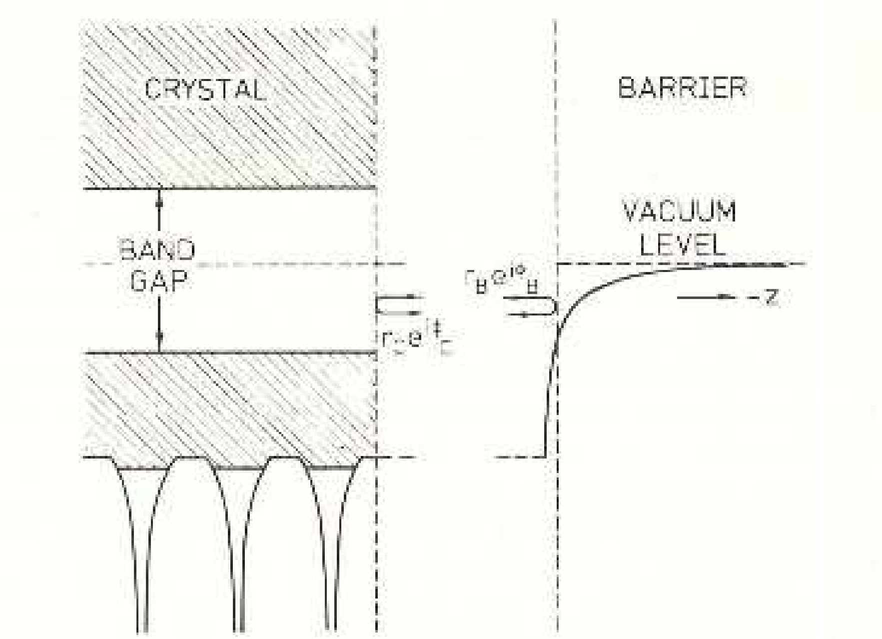
$$\phi_e^{SP}(\mathbf{r}) \equiv \langle \mathbf{r}|G_2^{-}|e,\mathbf{k}_{\parallel}\rangle$$

The semi-infinite Solid

Bulk potential



Surface potential



$\det|\mathbf{1} - \mathbf{R}_B\mathbf{R}_C| \;=\; 0 \quad \Rightarrow$ Surface state
$\det|\mathbf{1} - \mathbf{R}_B\mathbf{R}_C| \;=\; \min \Rightarrow$ Surface resonance

Dipole selection rules

Relativistic dipole selection rules

$M_{PES} \neq 0$ für $\boldsymbol{\Delta\kappa = \pm 1}$, $\kappa + \kappa' = \mathbf{0}$ und $\mu + \mu' = \mathbf{0}$

$$\mathcal{A}^{lm}_{\kappa'\mu'-\kappa''\mu''} = \int\limits_{(4\pi)} d\hat{\mathbf{r}}\ \chi^{\mu' t *}_{\kappa'}(\hat{\mathbf{r}})\ (\hat{\boldsymbol{\alpha}} \cdot \mathbf{A}_0)\ Y^m_l(\hat{\mathbf{r}})\ \chi^{\mu''}_{-\kappa''}(\hat{\mathbf{r}})$$

$\hat{\boldsymbol{\alpha}} \cdot \mathbf{A}_0$: relativistic form of electron-photon interaction

$$\chi^{\mu}_{\kappa}(\hat{\mathbf{r}}) = \sum_{s=\pm\frac{1}{2}} \mathcal{C}_{\kappa\mu s} Y^{\mu-s}_l(\hat{\mathbf{r}}) \chi^s$$

The spin-angular functions χ^{μ}_{κ} are given by the Pauli spinors χ^s, Clebsch-Gordan coefficients $\mathcal{C}_{\kappa\mu s}$ and by the spherical harmonics $Y^{\mu-s}_l(\hat{\mathbf{r}})$

Spinpolarized electronic structure for $\Sigma \neq 0$

$$\frac{\partial}{\partial r} C_{n\kappa'\mu'\kappa\mu}(r) = -pr^2 \left(n^u_{\kappa'}(kr) \mathcal{K}^+_{n\kappa'\mu'\kappa\mu}(r) + n^l_{\kappa'}(kr) \mathcal{K}^-_{n\kappa'\mu'\kappa\mu}(r) \right)$$

$$\frac{\partial}{\partial r} S_{n\kappa'\mu'\kappa\mu}(r) = -pr^2 \left(j^u_{\kappa'}(kr) \mathcal{K}^+_{n\kappa'\mu'\kappa\mu}(r) + j^l_{\kappa'}(kr) \mathcal{K}^-_{n\kappa'\mu'\kappa\mu}(r) \right), \qquad p = k \left(\frac{E + c^2}{c} \right)$$

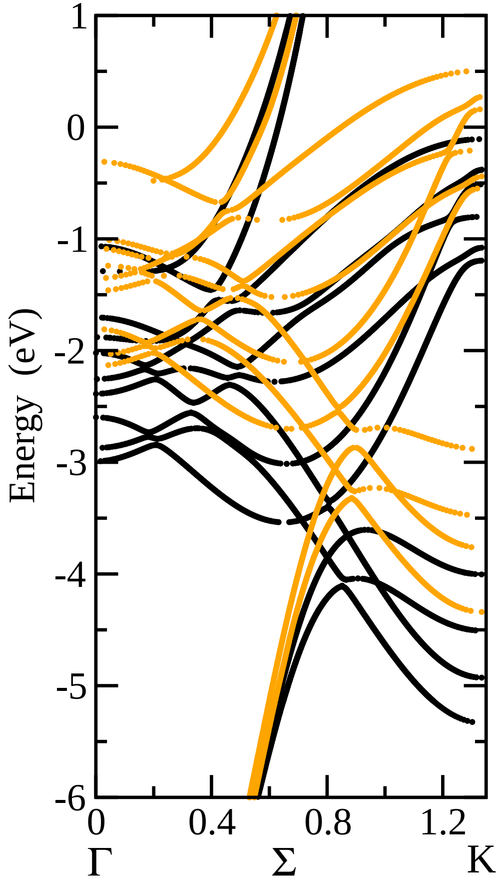
$$\mathcal{K}^+_{n\kappa'\mu'\kappa\mu}(r) = \sum_{\kappa'''\mu'''} \sum_{l''m''} \frac{1}{2} (U^{\uparrow}_{l''m''}(r) I^{+m}_{\kappa'\mu' l''m''\kappa'''\mu'''} + U^{\downarrow}_{l''m''}(r) I^{+p}_{\kappa'\mu' l''m''\kappa'''\mu'''}) \phi^u_{\kappa'''\mu'''\kappa\mu}(r)$$

$$\mathcal{K}^-_{n\kappa'\mu'\kappa\mu}(r) = \sum_{\kappa'''\mu'''} \sum_{l''m''} \frac{1}{2} (U^{\uparrow}_{l''m''}(r) I^{-p}_{\kappa'\mu' l''m''\kappa'''\mu'''} + U^{\downarrow}_{l''m''}(r) I^{-m}_{\kappa'\mu' l''m''\kappa'''\mu'''}) \phi^l_{\kappa'''\mu'''\kappa\mu}(r)$$

$$U^{\uparrow\downarrow}_{l''m''}(r, r', E) = V^{\uparrow\downarrow}(r) \delta(r - r') + \Sigma^{\uparrow\downarrow DMFT}_{l''m''}(E) \delta_{l''2}$$

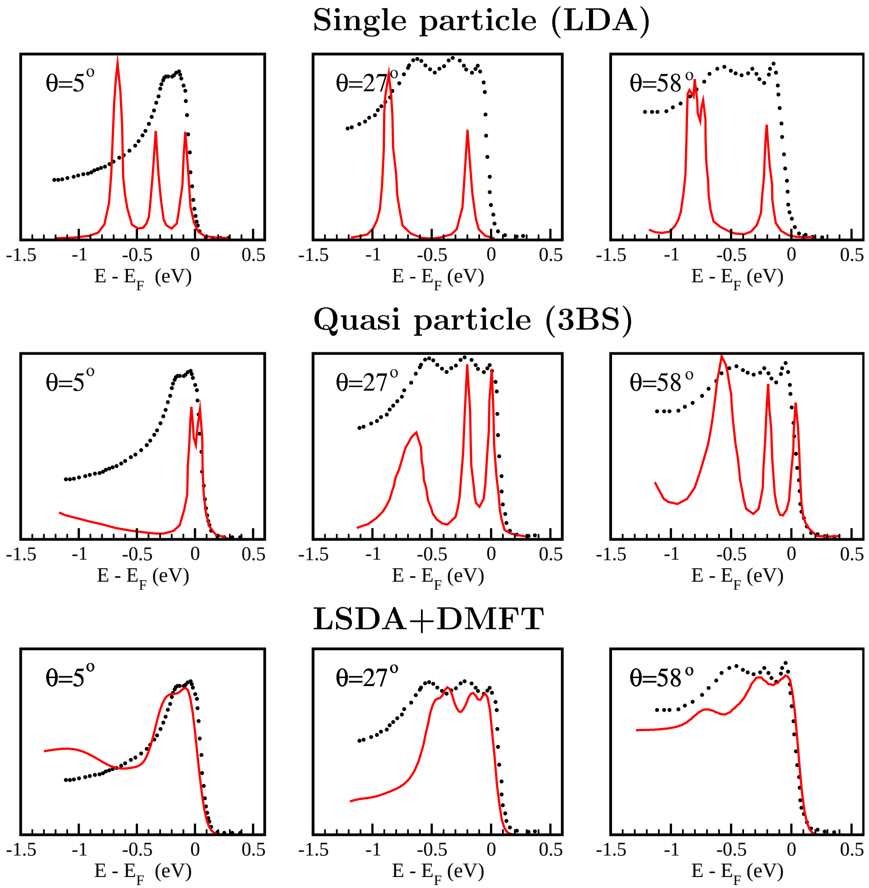
Ni(011): electronic structure and ARUPS

Spinpolarized bandstructure



Comparison between experiment and theory

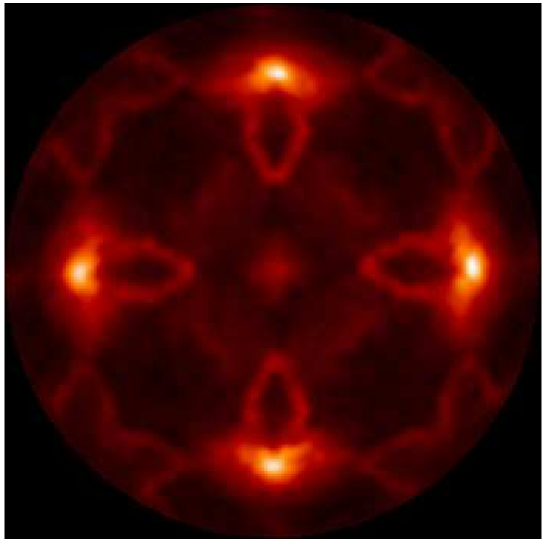
F. Manghi, J. Osterwalder et al. PRB 59, R10409 (1999)

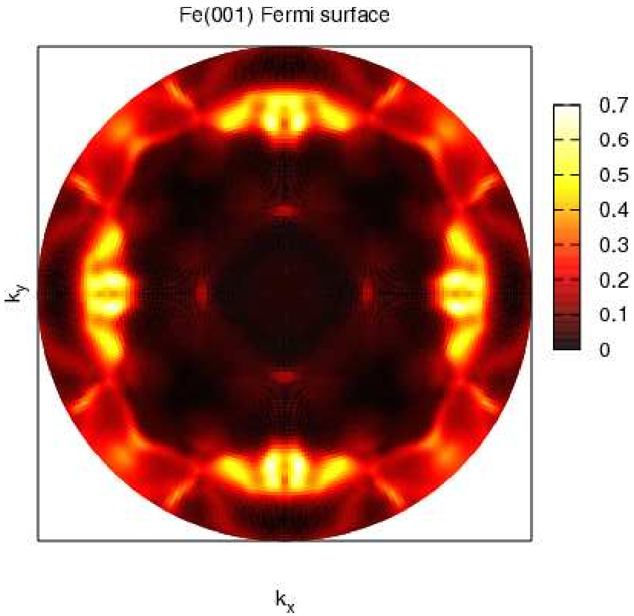


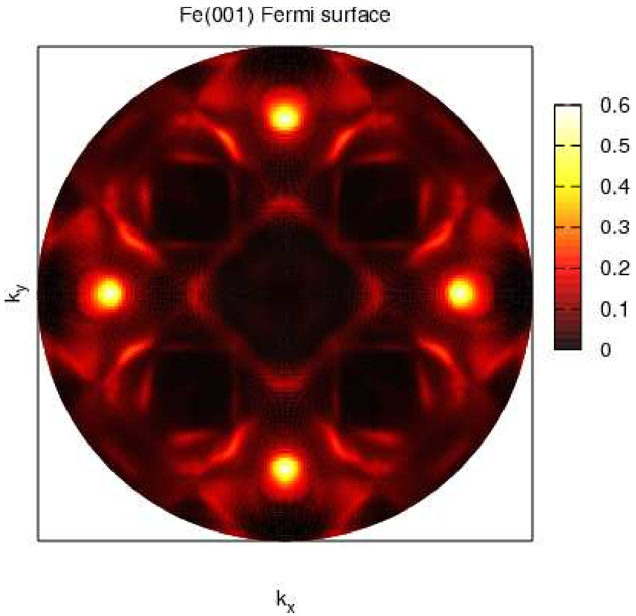
J. Braun, J. Minár et al., PRL 97, 227601 (2006)

Fe(001): Fermi surface

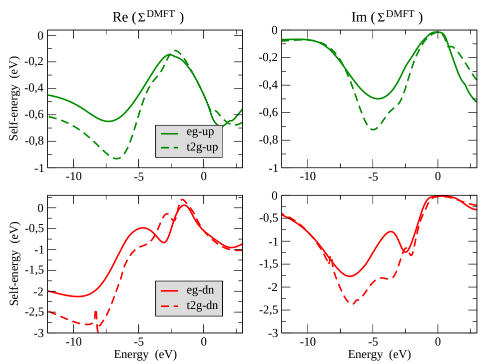
Experiment | Theory LSDA | Theory LSDA+DMFT





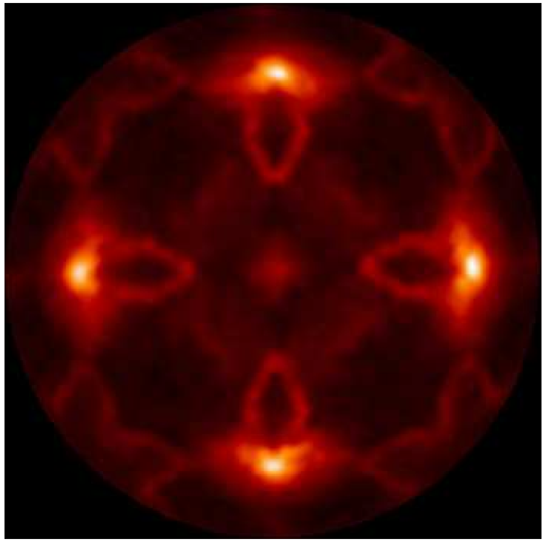


- $E_{h\nu}$=50eV, linear polarised light
- Experiments Mulazzi et al.

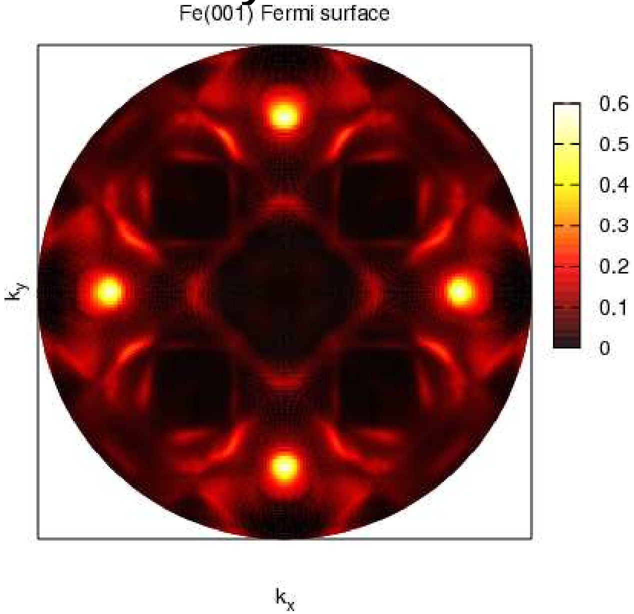


Fe(001): Fermi surface

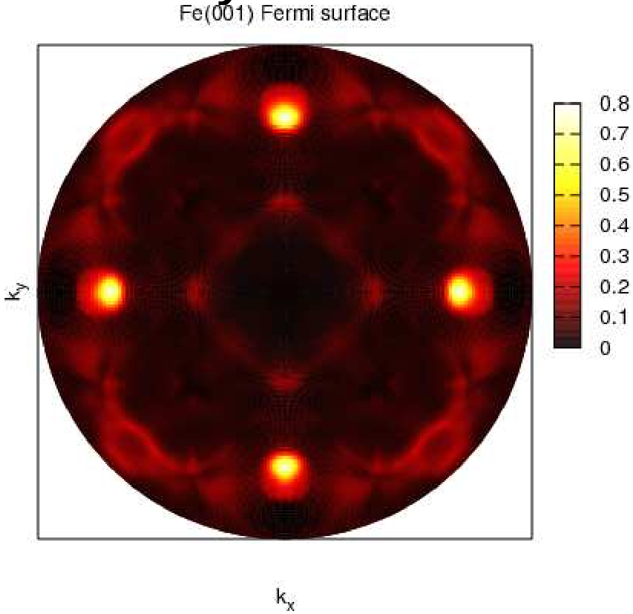
Experiment



Theory surface

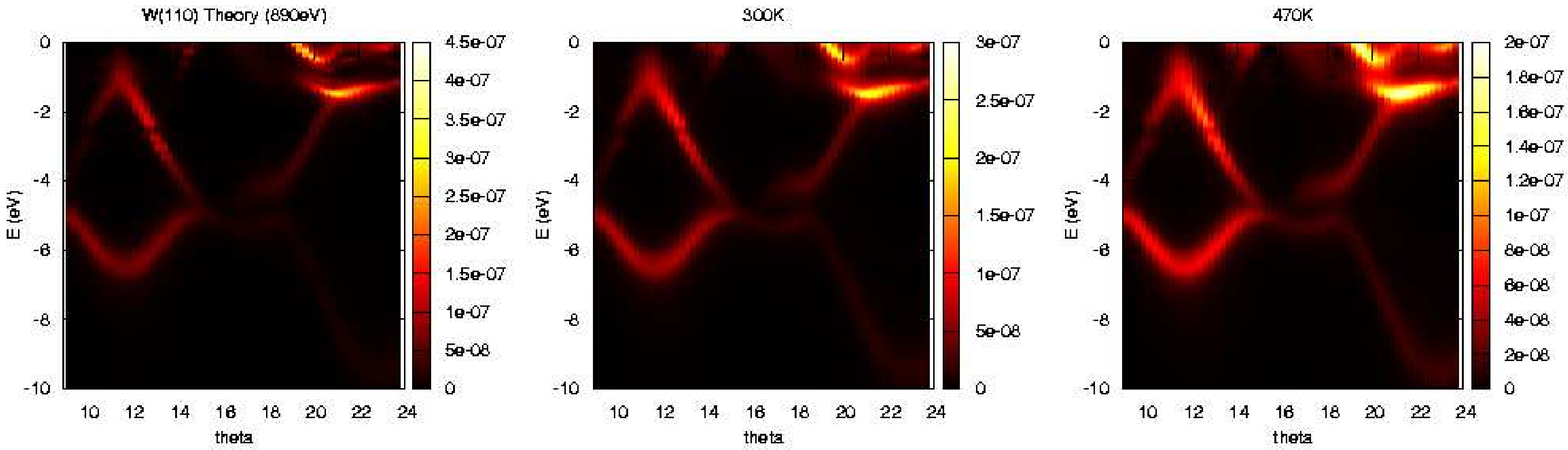


Theory bulk

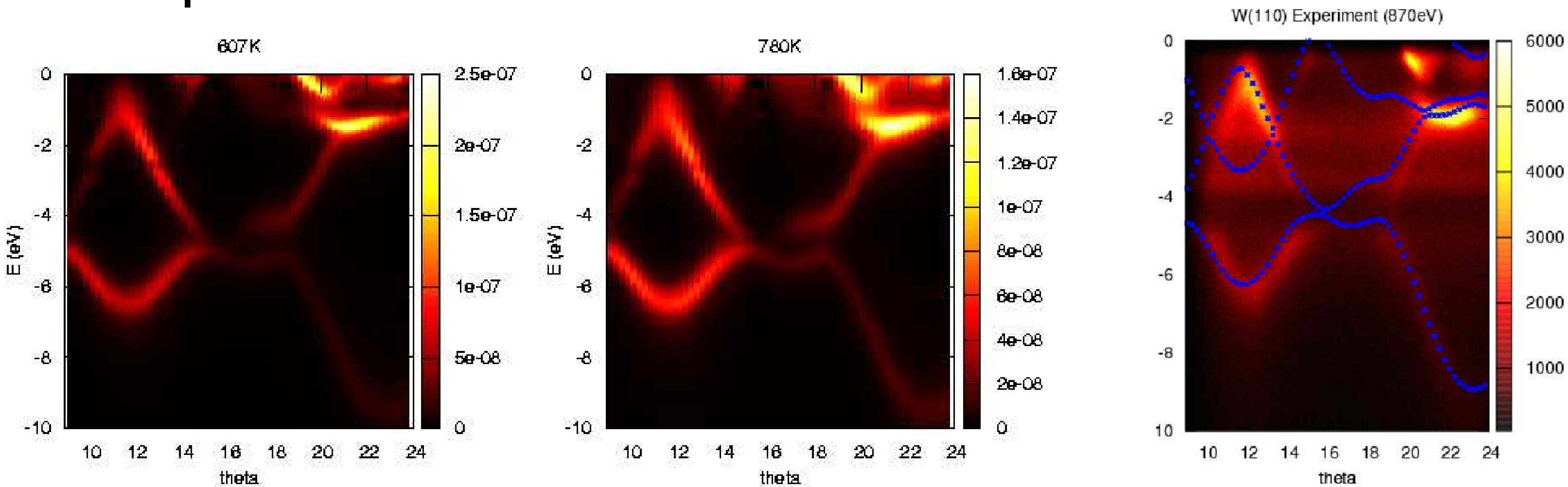


- $E_{h\nu}$=50eV, p-polarised light
- Experiments Mulazzi et al.

W(011): X-ray photoemission at T $\neq$ 0 K^o



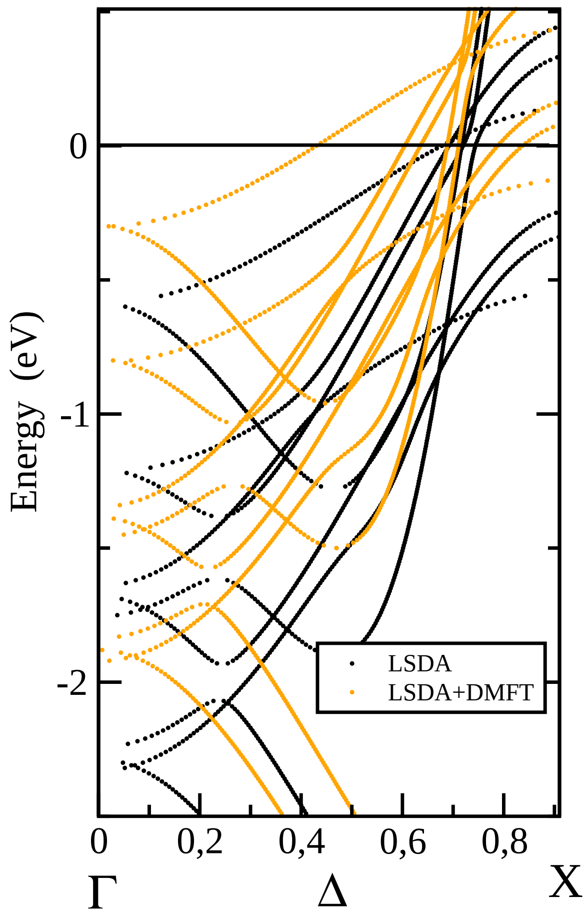
- effect of photon momentum considered



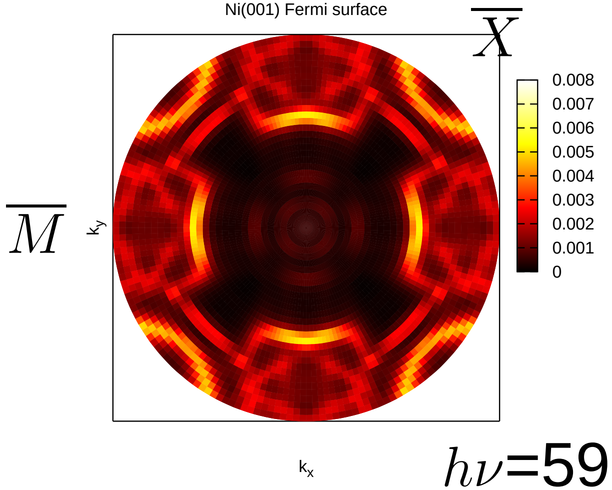
- Temperature-dependent PE
- within Debey-Waller model
- $E_{h\nu}$=870eV, T=780 K
- Exp. Plucinski et al.

Ni(001): X-ray photoemission and Fermi surface

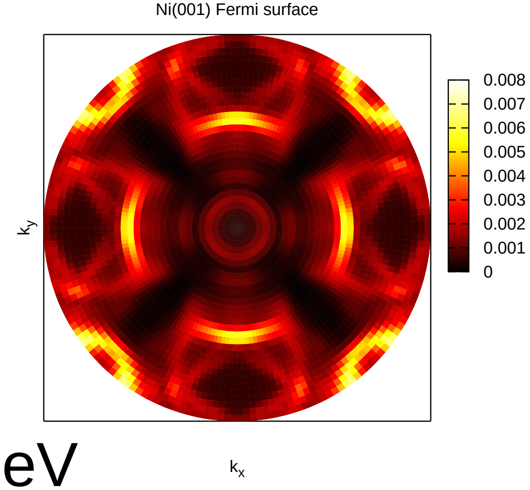
Spinpolarised bandstructure



LSDA

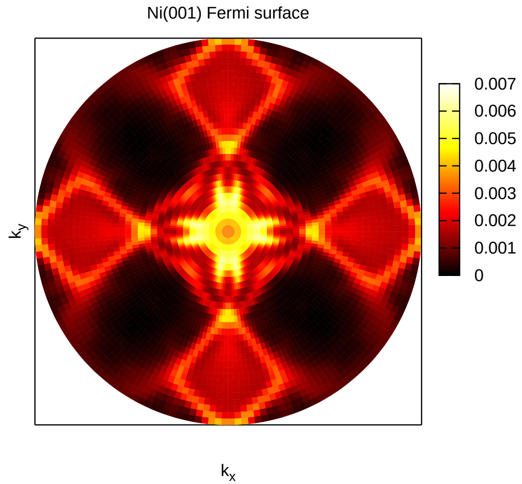


LSDA+DMFT



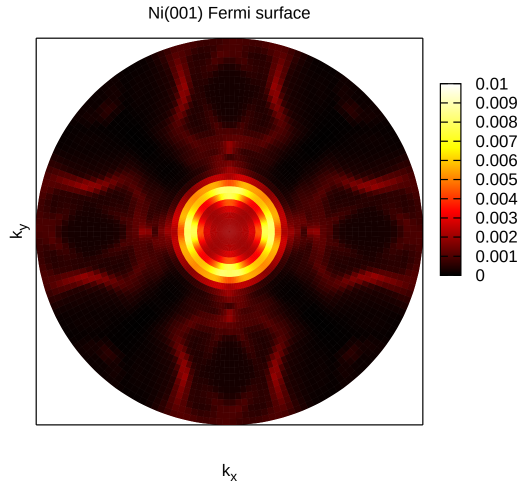
$h\nu$=595 eV

LSDA+DMFT



$h\nu$=700 eV

LSDA+DMFT



$h\nu$=780 eV

Conclusions and Outlook

Relativistic DFT-ONE-STEP-DMFT (REDOSD) approach

$\Downarrow$

Improved description of the electronic structure of ferromagnetic 3d-metals
Detailed analysis of electronic dispersion in the UV- and X-ray regime

Spectroscopic investigations on Mn and hexagonal Co, NiO or FeCo

Consideration of the spatial dependence of the electronic self-energy Σ

$\Downarrow$

Numerical solution of the corresponding integro-differential equation for Σ

$\Downarrow$

Spectroscopic investigations to highly correlated materials and consideration of Phonon effects beyond Debey-Waller

Combination of the TBKKR-method with the REDOST approach

$\Downarrow$

Quantitative description of electronic structure and photoemission from arbitrary 2D-structures like multilayers or adsorbate systems

System of radial Dirac equations

Magnetic couplings

MC $\neq 0$ for $B_z : \delta_{\kappa,\kappa'(-\kappa'-1)}\delta_{\mu,\mu'}$ $B_{xy} : \delta_{\kappa,\kappa'(-\kappa'-1)}\delta_{\mu,\mu'\pm1}$

$\kappa = l,\ \kappa > 0;\ \ \kappa = -l-1,\ \kappa < 0;\ \ \mu = l + s = -|\kappa| + 0.5, ..., |\kappa| - 0.5$

$$\phi^{\mu}_{n\kappa}(\mathbf{r}) = \sum_{\kappa'\mu'} J^{\mu'}_{\kappa'}(\mathbf{r})C_{n\kappa\mu\kappa'\mu'}(r) - N^{\mu'}_{\kappa'}(\mathbf{r})S_{n\kappa\mu\kappa'\mu'}(r)$$

$$\frac{\partial}{\partial r}C_{n\kappa'\mu'\kappa\mu}(r) = -pr^2\left(n^u_{\kappa'}(kr)\mathcal{K}^+_{n\kappa'\mu'\kappa\mu}(r) + n^l_{\kappa'}(kr)\mathcal{K}^-_{n\kappa'\mu'\kappa\mu}(r)\right)$$

$$\frac{\partial}{\partial r}S_{n\kappa'\mu'\kappa\mu}(r) = -pr^2\left(j^u_{\kappa'}(kr)\mathcal{K}^+_{n\kappa'\mu'\kappa\mu}(r) + j^l_{\kappa'}(kr)\mathcal{K}^-_{n\kappa'\mu'\kappa\mu}(r)\right), \qquad p = k\left(\frac{E+c^2}{c}\right)$$

$$\mathcal{K}^+_{n\kappa'\mu'\kappa\mu}(r) = \sum_{\kappa'''\mu'''}\sum_{l''m''}\frac{1}{2}(V^{up}_{nl''m''}(r)I^{+m}_{\kappa'\mu'l''m''\kappa'''\mu'''} + V^{down}_{nl''m''}(r)I^{+p}_{\kappa'\mu'l''m''\kappa'''\mu'''})\phi^u_{n\kappa'''\mu'''\kappa\mu}(r)$$

$$\mathcal{K}^-_{n\kappa'\mu'\kappa\mu}(r) = \sum_{\kappa'''\mu'''}\sum_{l''m''}\frac{1}{2}(V^{up}_{nl''m''}(r)I^{-p}_{\kappa'\mu'l''m''\kappa'''\mu'''} + V^{down}_{nl''m''}(r)I^{-m}_{\kappa'\mu'l''m''\kappa'''\mu'''})\phi^l_{n\kappa'''\mu'''\kappa\mu}(r)$$

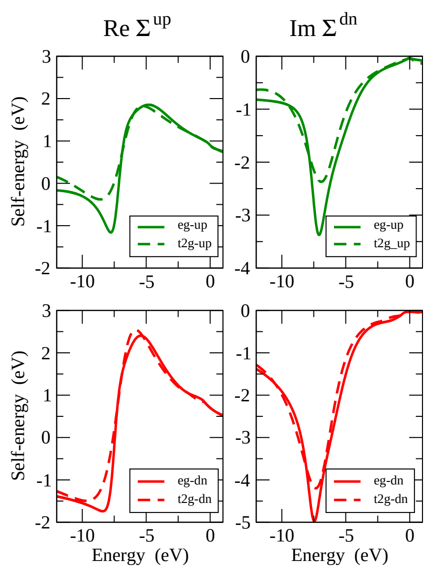
The integrals $I^{\pm p/m}$ can be written in terms of Clebsch-Gordan coefficients $C_{\kappa,\mu,s}$, multiplied by the three components of the direction vector $\mathbf{b}$ of $\mathbf{B}$ and by the Gaunt coefficients $I_{l'm'l''m''l'''m'''}$, which describe the angular mixing.

Ni(001): electronic structure and ARUPS

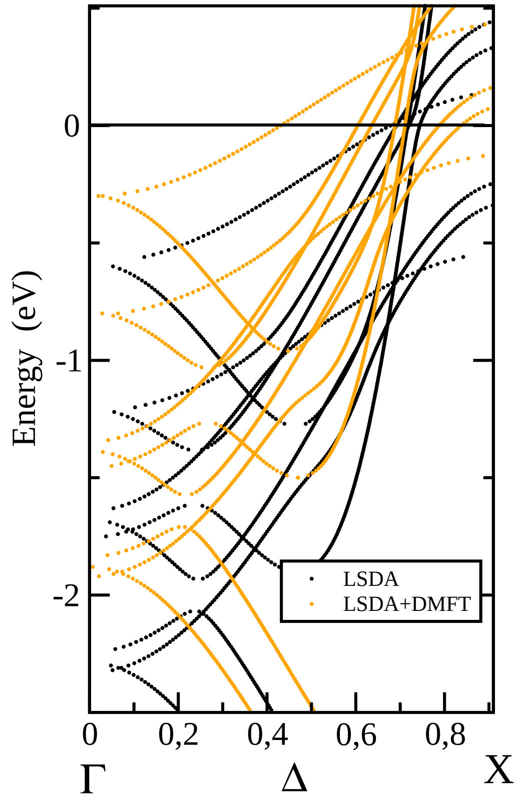
Non-local potential U

$$U_{lm}^{\uparrow\downarrow}(r, r', E) =$$

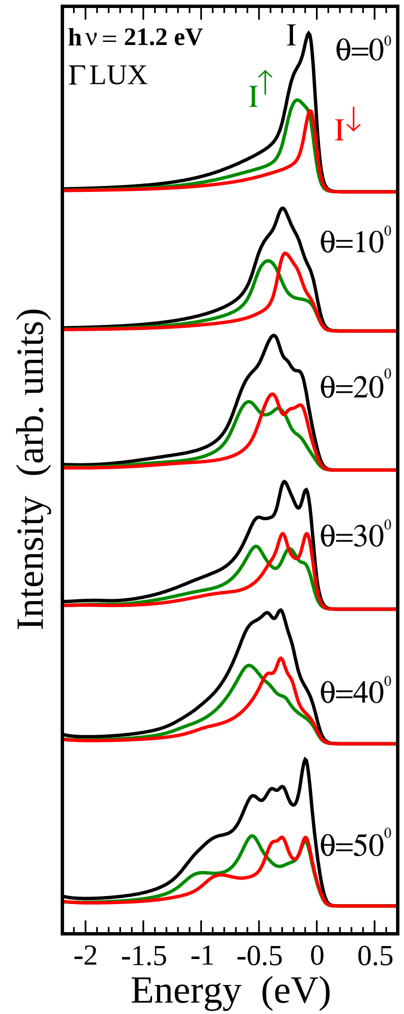
$$V^{\uparrow\downarrow}(r)\delta(r-r') + \Sigma_{lm}^{\uparrow\downarrow DMFT}(E)\delta_{l2}$$



Spinpolarised bandstructure

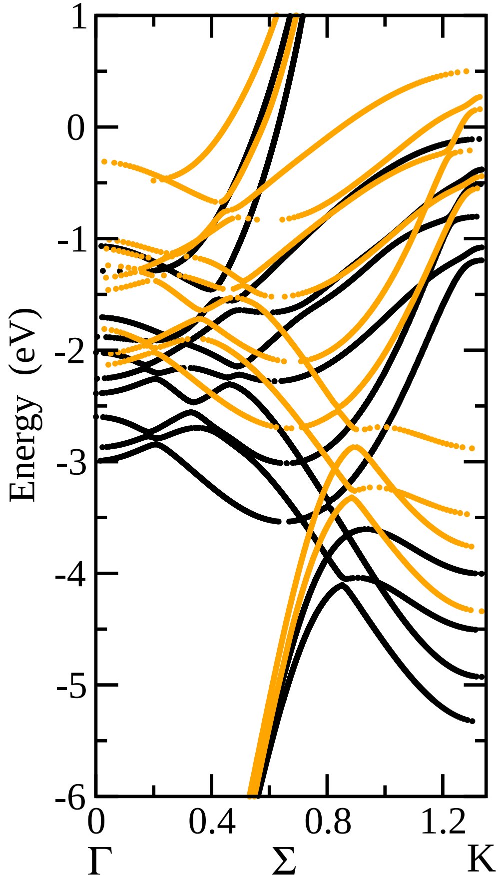


Off-normal emission: $\overline{\Gamma X}$



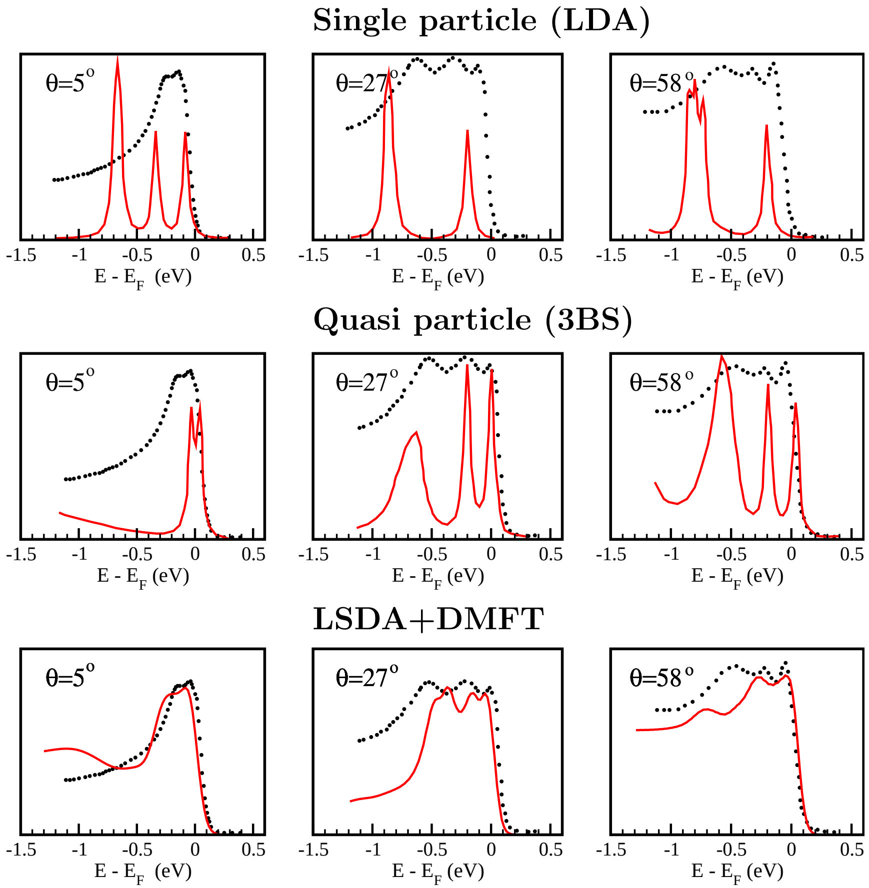
Ni(011): electronic structure and ARUPS

Spinpolarized bandstructure



Comparison between experiment and theory

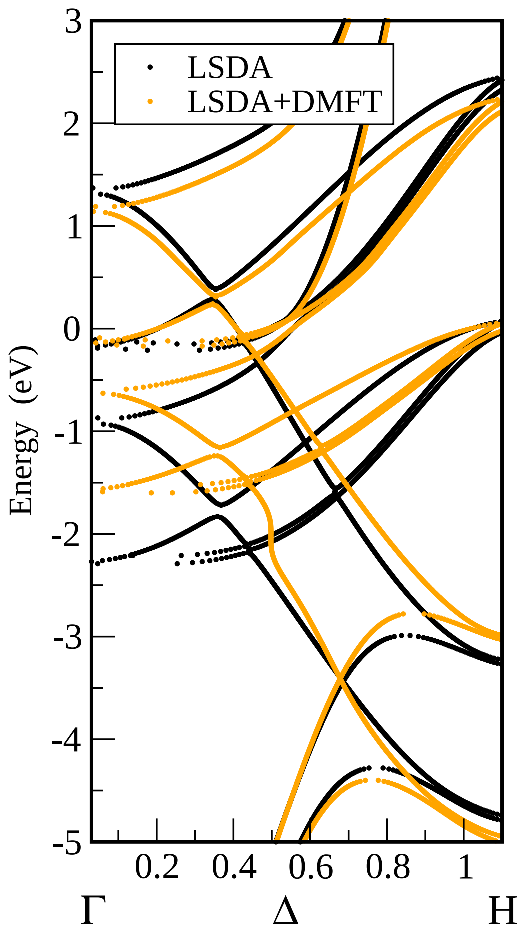
F. Manghi, J. Osterwalder et al. PRB 59, R10409 (1999)



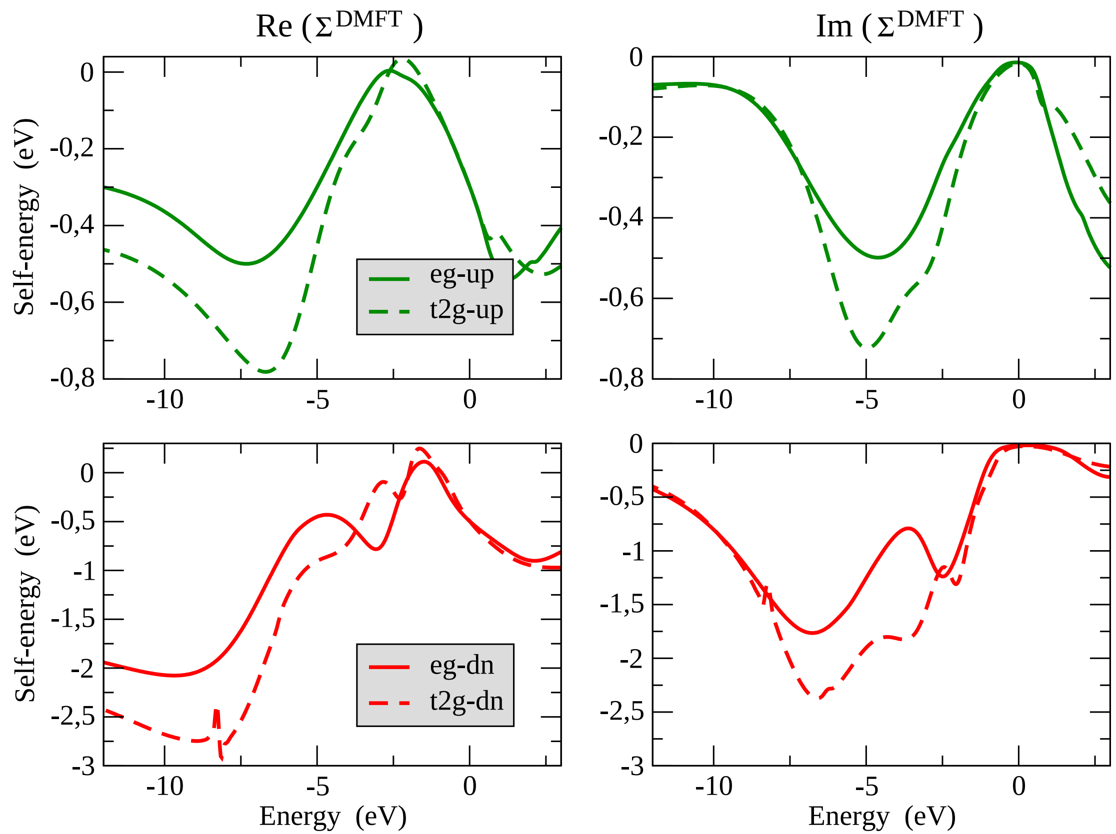
J. Braun, J. Minár et al., PRL 97, 227601 (2006)

Fe(001): electronic structure

Spinpolarized bandstructure



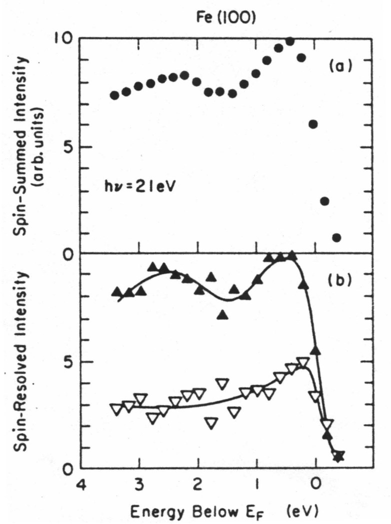
Spin- and symmetry resolved self-energy for Fe

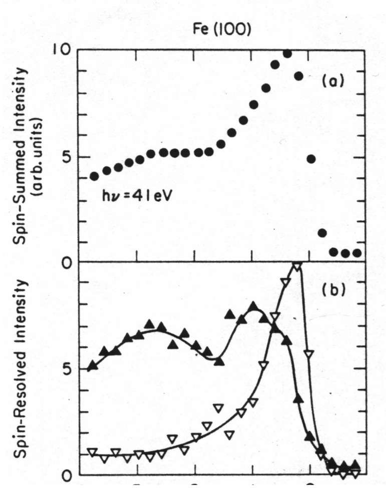


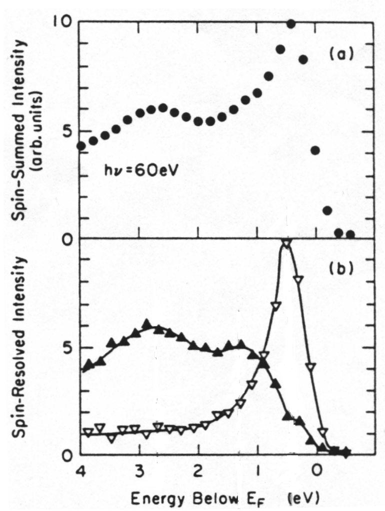
Fe(001): ARUPS

Comparison between experiment and theory

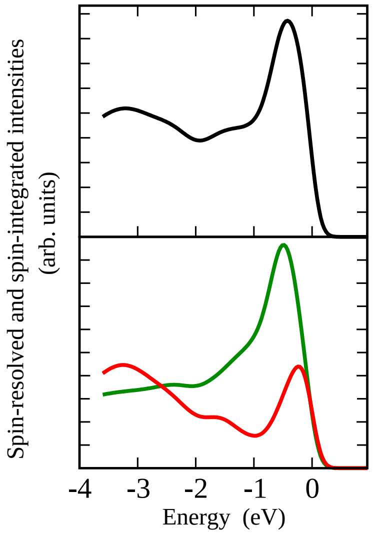
E. Kisker et al. PRB 31 329 (1984)

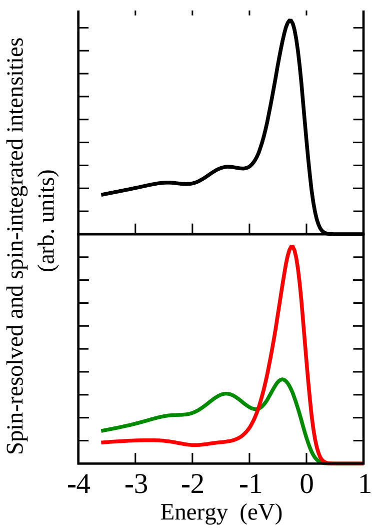


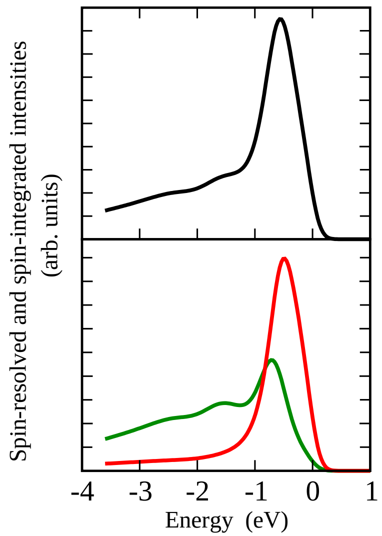




J. Braun, J. Minár and H. Ebert

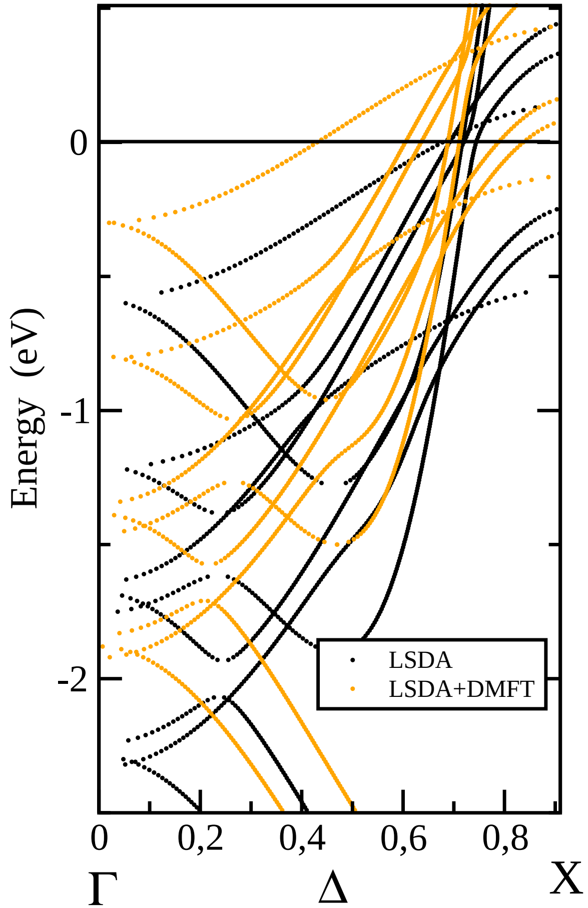




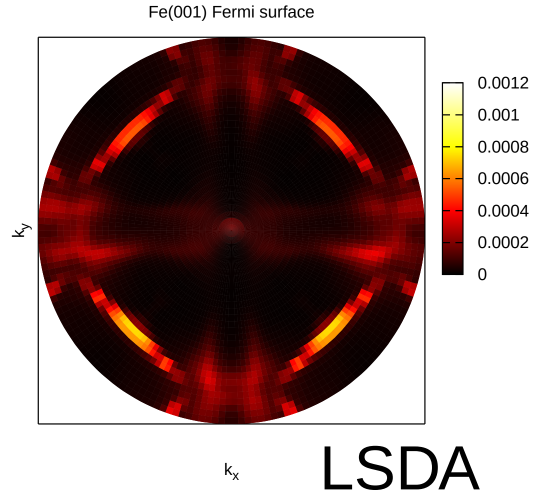


Ni(001): X-ray photoemission

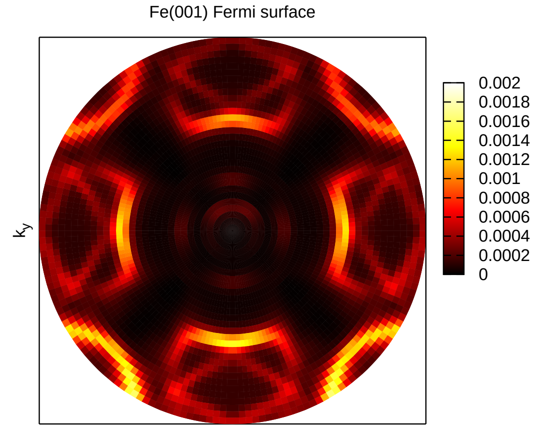
Spinpolarised bandstructure



Majority spin

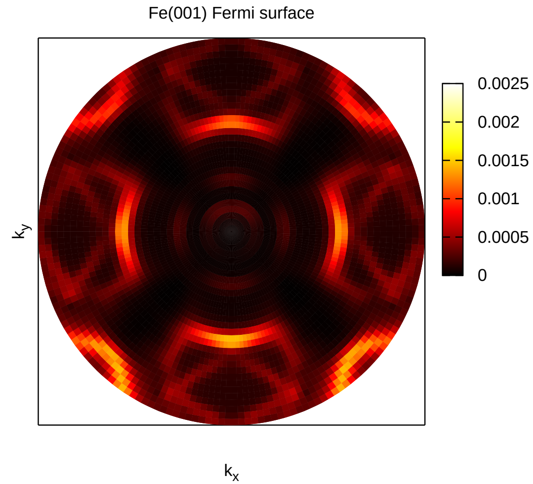


Minority spin

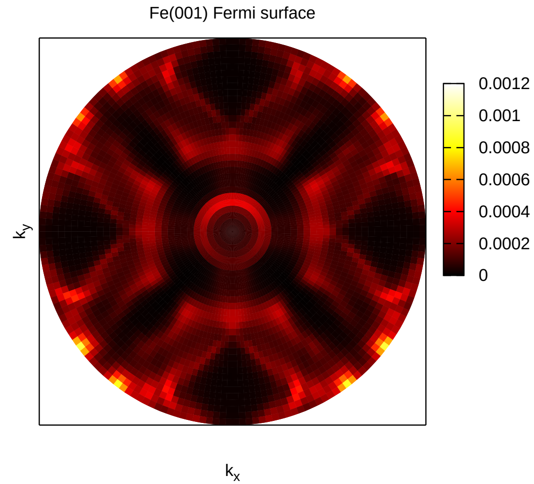


LSDA $h\nu$=595 eV

Majority spin



Minority spin



LSDA+DMFT $h\nu$=595 eV