KKR+DMFT/Spectroscopies

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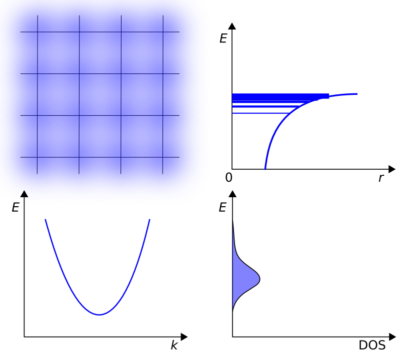
Outline

- origin of localization
- local electron interactions
  - statics: LDA+U
  - dynamics: LDA+DMFT
- applications to spectroscopies
  - optics and magneto-optics
  - x-ray photoemission
  - x-ray absorption and magnetic circular dichroism

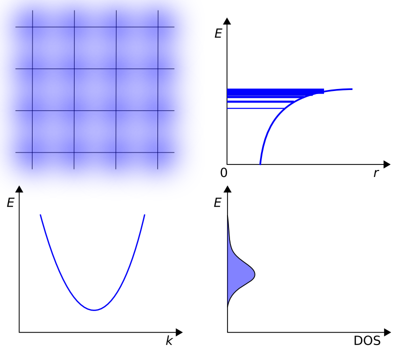
Localized electrons



$$-\frac{\hbar^2}{2m}\nabla^2\psi(\vec{r}) + V(\vec{r})\psi(\vec{r}) = E\psi(\vec{r})$$

$$\psi(\vec{r}) = \sum_{lm}\left(\frac{u_l(r)}{r}\right)Y_{lm}(\hat{r})$$

Localized electrons



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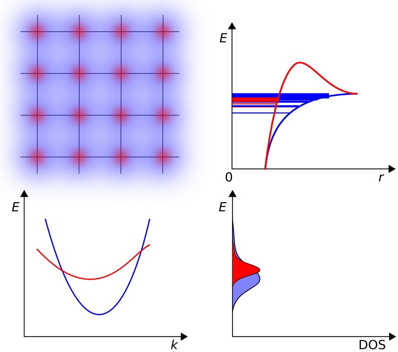
$$-\frac{\hbar^2}{2m}\frac{d^2u_l}{dr^2} + \left(V + \underbrace{\frac{\hbar^2}{2m}\frac{l(l+1)}{r^2}}_{\text{centrifugal potential}}\right)u_l = Eu_l$$

s-electrons: $V_{\mathrm{cf}} \sim 0$
p-electrons: $V_{\mathrm{cf}} \sim 2$

Localized electrons



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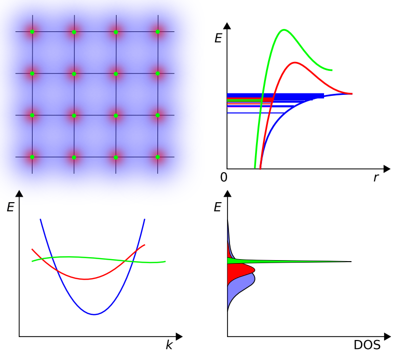
d-electrons: $V_{\mathrm{cf}} \sim 6$

Localized electrons



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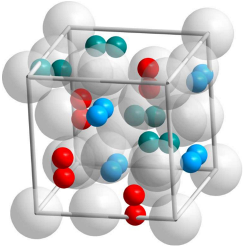
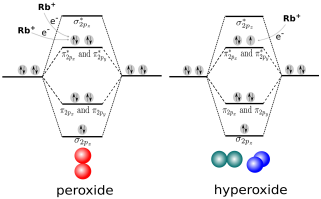
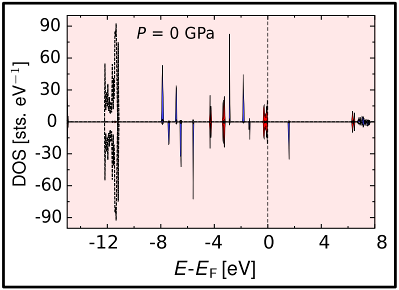
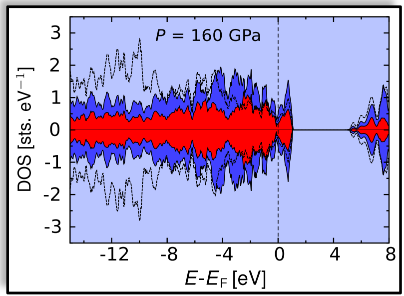
d-electrons: $V_{\mathrm{cf}} \sim 6$
f-electrons: $V_{\mathrm{cf}} \sim 12$

Localized electrons: Rb_4O_6



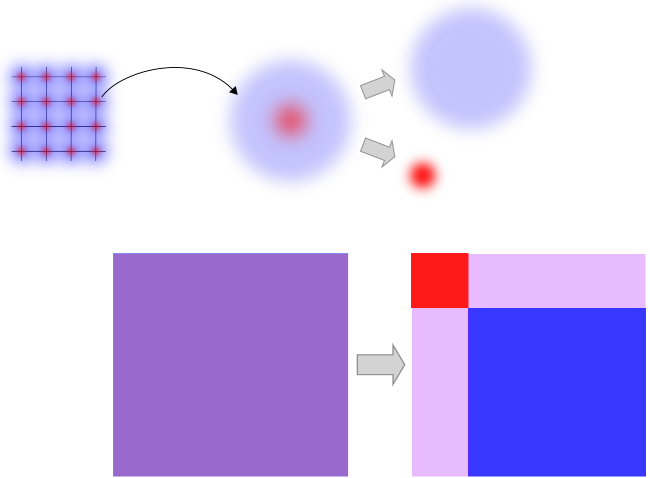




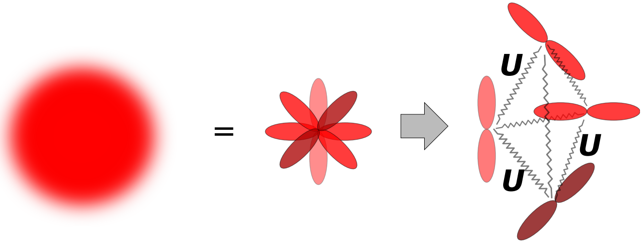


S. Naghavi, *et al*, PRB 2011

Basis decomposition



Inside effective impurity

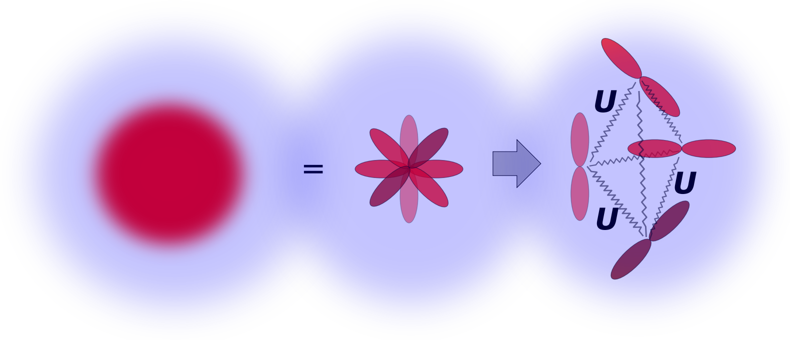


$$U_{ij} = \int dr_1 dr_2 \frac{\psi_i^*(r_1)\psi_i(r_1)\psi_j^*(r_2)\psi_j(r_2)}{|r_1 - r_2|} + \int dr_1 dr_2 \frac{\psi_i^*(r_1)\psi_j(r_1)\psi_j^*(r_2)\psi_i(r_2)}{|r_1 - r_2|}$$

$$U_{ijkl} = \int dr_1 dr_2 \frac{\psi_i^*(r_1)\psi_j(r_1)\psi_k^*(r_2)\psi_l(r_2)}{|r_1 - r_2|}$$

$$\psi_i(\vec{r}) = R_i(r)Y_i(\hat{r}) \Longrightarrow U_{ijkl} = \underbrace{U_R}_{\underbrace{F_0}_{U},\underbrace{F_2,F_4,}_{U-J,J}..} \cdot \underbrace{\delta(ijkl)}_{\text{ang. symmetry}}$$

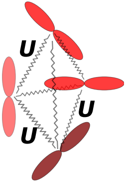
Inside effective impurity



$$U_{\mathrm{eff}}\ (\sim\ 3\ \mathrm{eV}) < U\ (\sim\ 10\ \mathrm{eV})$$

First step: LDA+U

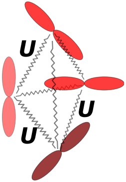
simple picture:



$$E_{\rm tot} = E_{\rm tot}^{\rm LDA} + {}^1\!/_2\, U \sum_{i\neq j} n_i n_j - \underbrace{{}^1\!/_2\, UN(N-1)}_{\rm db-c.}$$

$$\epsilon_i = \frac{\partial E_{\rm tot}}{\partial n_i} = \epsilon^{\rm LDA} + U\left(\frac{1}{2} - n_i\right)$$

simple picture:



$$E_{\text{tot}} = E_{\text{tot}}^{\text{LDA}} + {}^{1}\!/_{2}\, U \sum_{i \neq j} n_i n_j - \underbrace{{}^{1}\!/_{2}\, UN(N-1)}_{\text{db-c.}}$$

$$\epsilon_i = \frac{\partial E_{\text{tot}}}{\partial n_i} = \epsilon^{\text{LDA}} + U \left(\frac{1}{2} - n_i \right)$$

more strictly:

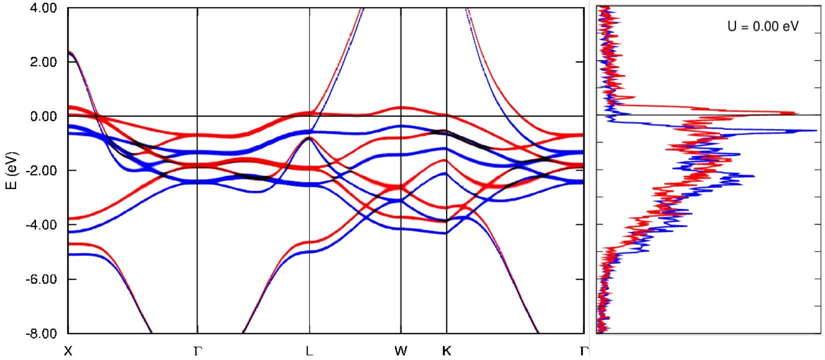
$$\sum_{1234} V_{1234} \cdot n_{12} n_{34} - \sum_{1234} V_{1234} \cdot \overbrace{\langle n_{12} n_{34} \rangle}^{n_{12}\langle n_{34}\rangle + \langle n_{12}\rangle n_{34} - \langle n_{12}\rangle\langle n_{34}\rangle}$$

$$\Rightarrow \frac{\partial}{\partial n_{12}} \Rightarrow \sum_{34 \neq 12} V_{1234} \left(n_{34} - \langle n_{34} \rangle \right)$$

what means $\langle\rangle$? $\quad \langle n \rangle_{\nu} = \frac{1}{2} \sum_{\sigma} n_{\sigma\nu}, \qquad \langle n \rangle_{\sigma} = \frac{1}{N} \sum_{\nu} n_{\sigma\nu}$

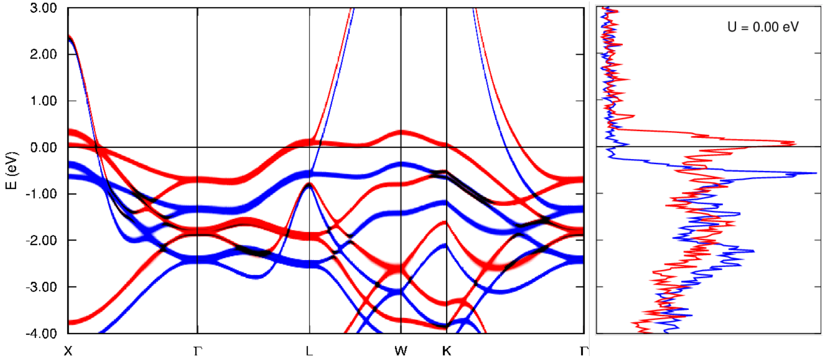
Ni: LDA+U (spin polarization)

$$\epsilon_{\nu\sigma} = \epsilon_{\nu\sigma}^{\mathrm{LDA}} + \sum_{\sigma'\nu' \neq \sigma\nu} V_{\sigma\nu\sigma'\nu'} \left(n_{\nu'\sigma'} - \langle n \rangle_\sigma \right)$$

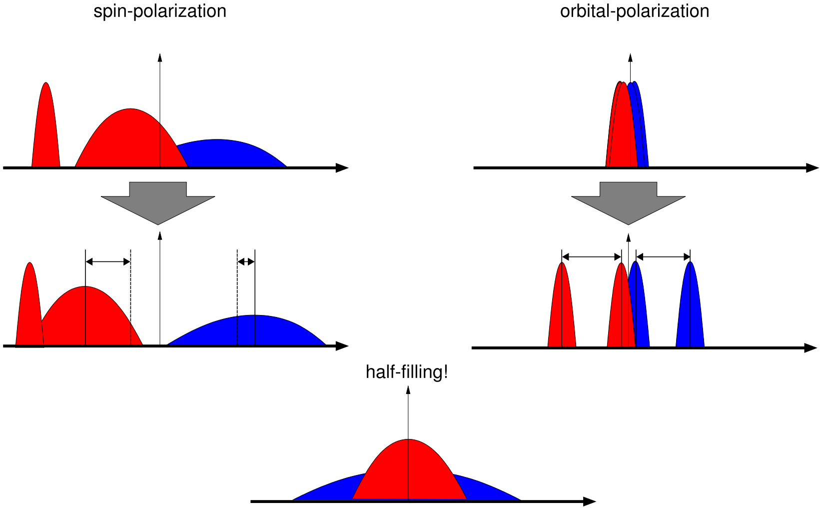


Ni: LDA+U (orbital polarization)

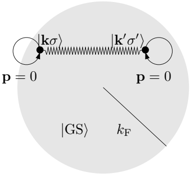
$$\epsilon_{\nu\sigma} = \epsilon_{\nu\sigma}^{\mathrm{LDA}} + \sum_{\sigma'\nu' \neq \sigma\nu} V_{\sigma\nu\sigma'\nu'} \left(n_{\nu'\sigma'} - \langle n \rangle_{\nu} \right)$$



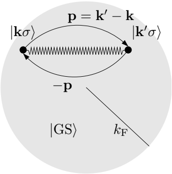
Half-filling



First-order processes



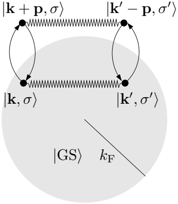
direct interaction



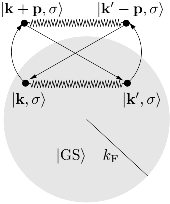
exchange interaction

$$E^{(1)} \sim \langle \mathrm{FS}\,|V|\,\mathrm{FS}\rangle$$

Second-order processes



direct interaction

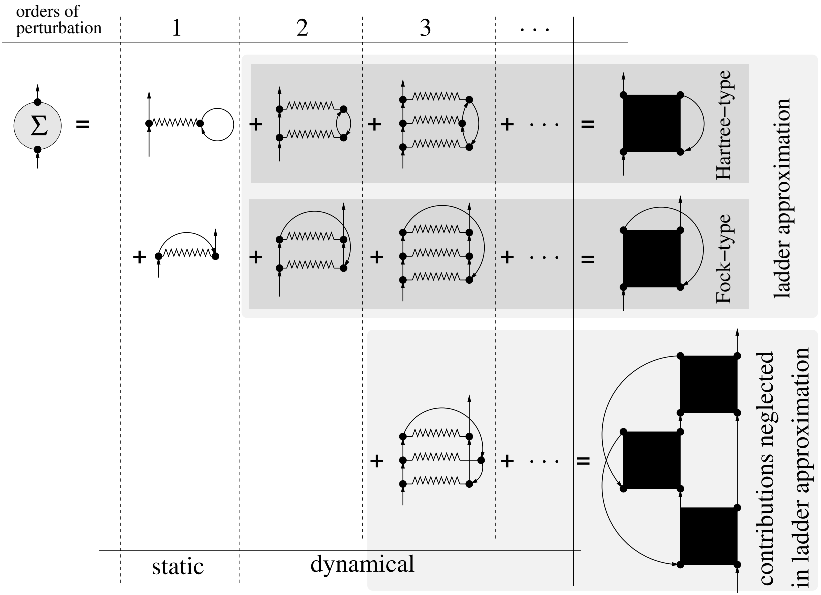


exchange interaction

$$E^{(2)} \sim \frac{\langle \mathrm{FS}|V|\nu\rangle\langle\nu|V|\mathrm{FS}\rangle}{E_0 - E_\nu}$$

- A. L. Fetter, J. D. Walecka,
  Quantum Theory of Many-Particle Systems
- L. Kadanoff, G. Baym,
  Quantum Statistical Mechanics

AIM solver

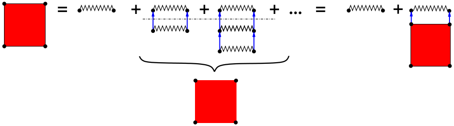


orders of perturbation
1
2
3
. . .
Σ =
+
+
+
+ . . .
=
Hartree–type
Fock–type
ladder approximation
contributions neglected in ladder approximation
static
dynamical

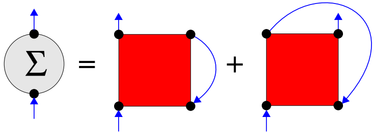
Self-energy

$$T_{1234}(\epsilon) = V_{1234} + \sum{}' V_{121'2'}\, \Phi_{1'2'3'4'}(\epsilon)\, T_{3'4'34}(\epsilon)$$

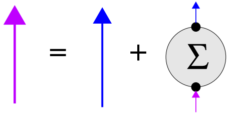
$$\Phi_{1234}(\epsilon) = i \int \frac{d\epsilon'}{2\pi} G_{13}(\epsilon - \epsilon') G_{24}(\epsilon')$$



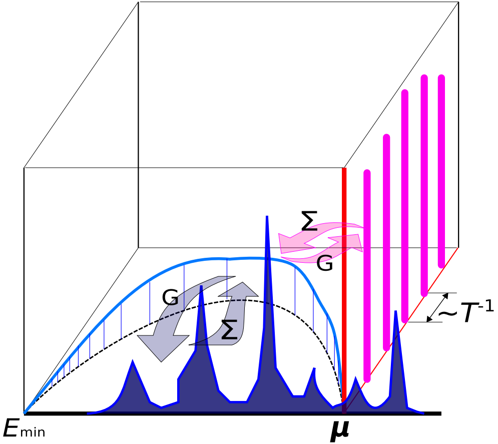
$$\Sigma_{12}(\epsilon) = \frac{1}{i} \int \frac{d\epsilon'}{2\pi} \left[T_{121'2'}(\epsilon' + \epsilon) - T_{211'2'}(\epsilon' + \epsilon) \right] G_{2'1'}(\epsilon')$$



$$G^{-1}(\epsilon) = G^{-1}(\epsilon) + \Sigma(\epsilon)$$



Self-energy



- Real-energy axis: $T = 0$

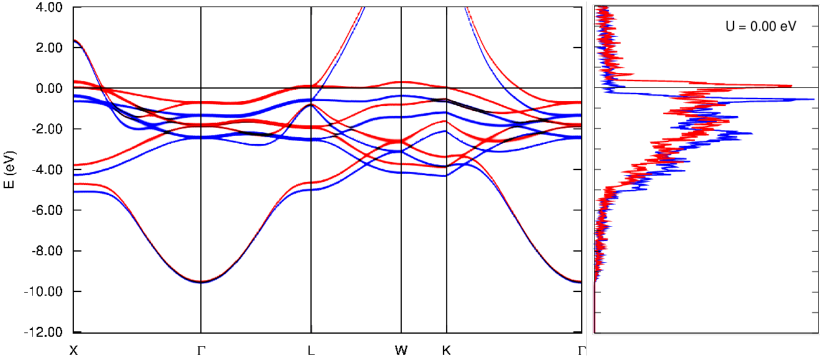
$$\int_{-\infty}^{\infty} \Phi(\epsilon)\Theta(\epsilon;\mu,0)d\epsilon = \int_{-\infty}^{\mu} \Phi(\epsilon)d\epsilon$$

- Matsubara poles: $T > 0$

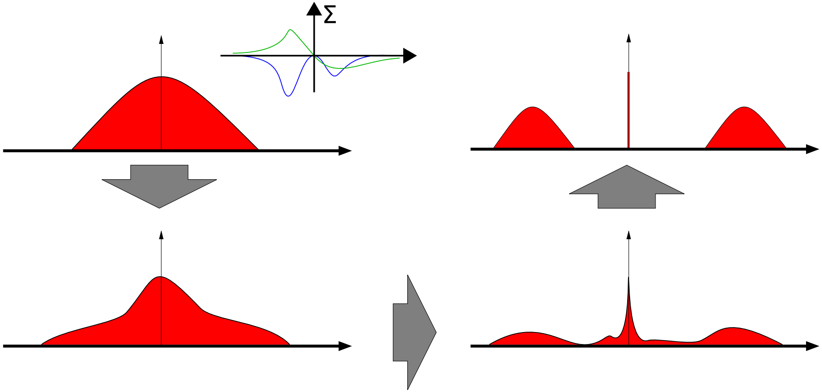
$$\int_{-\infty}^{\infty} \Phi(\epsilon)\Theta(\epsilon;\mu,T)d\epsilon = \sum_k \Phi\left(\frac{(2k+1)\pi}{k_{\rm B}T}\right)$$

Ni: LDA+DMFT

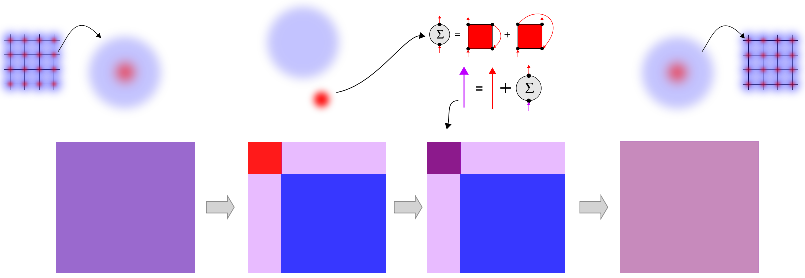
$$G^{-1}(\epsilon) = G^{-1}(\epsilon) + \Sigma(\epsilon)$$



Metal/Mott insulator



Coupling with external bath



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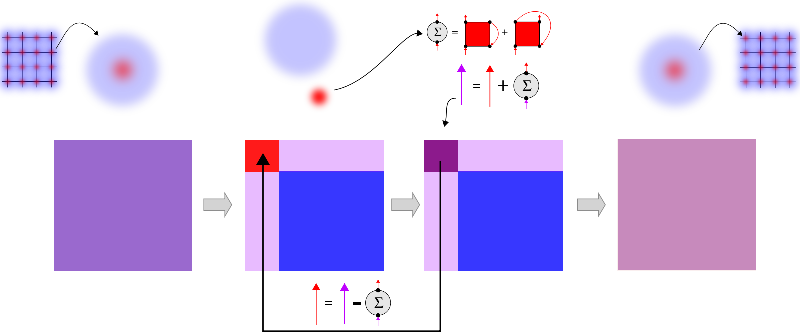
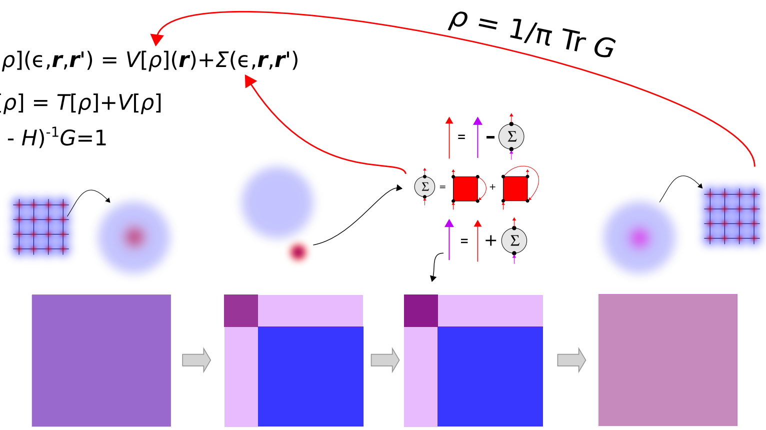


Coupling with external bath

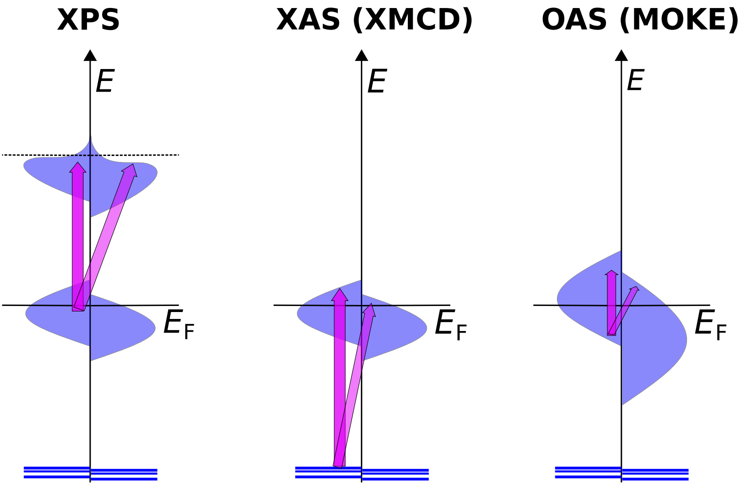
$V[\rho](\epsilon,\boldsymbol{r},\boldsymbol{r}') = V[\rho](\boldsymbol{r})+\Sigma(\epsilon,\boldsymbol{r},\boldsymbol{r}')$

$H[\rho] = T[\rho]+V[\rho]$

$(\epsilon - H)^{-1}G=1$



Spectroscopies



Fermi "golden rule" and Green's functions

transition probability from $\langle m|$ to $\langle n|$:

$$\sim |\langle m| X |n\rangle|^2 = \langle m| X |n\rangle \langle n| X |m\rangle$$

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the whole intensity:

$$\sum_{nm} \langle m| X |n\rangle \langle n| X |m\rangle \Rightarrow \sum_{nm} \langle m| X |n\rangle \langle n| X |m\rangle \Theta(\epsilon_n - \mu)\Theta(\mu - \epsilon_m)\delta(\epsilon_n - \epsilon_m - \omega)$$

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$$= \sum_{nm} \langle m| X |n\rangle \langle n| X |m\rangle \int_{\mu}^{\mu+\omega} d\epsilon \frac{1}{(\epsilon - \epsilon_n)(\epsilon - \omega - \epsilon_m)}$$

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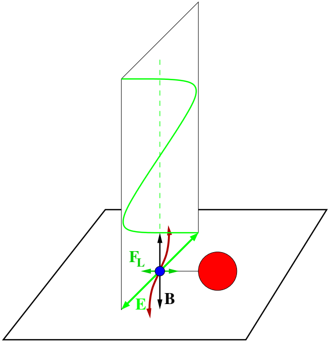
$$= \int_{\mu}^{\mu+\omega} d\epsilon \, X \sum_{n} \frac{|n\rangle \langle n|}{\epsilon - \epsilon_n} X \sum_{m} \frac{|m\rangle \langle m|}{\epsilon - \omega - \epsilon_m} = \int_{\mu}^{\mu+\omega} d\epsilon \, X \, G(\epsilon) \, X \, G(\epsilon - \omega)$$

MOKE

$$\theta_{\mathrm{K}}(\omega) + \mathrm{i}\varepsilon_{\mathrm{K}}(\omega) = \frac{-\sigma_{\mathrm{xy}}(\omega)}{\sigma_{\mathrm{xx}}(\omega)\sqrt{1 + \frac{4\pi \mathrm{i}}{\omega}\sigma_{\mathrm{xx}}(\omega)}}$$



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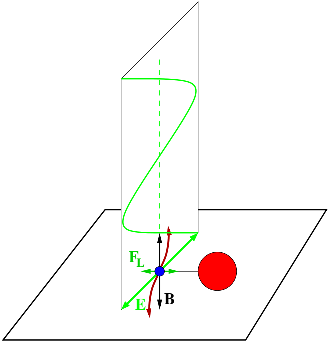

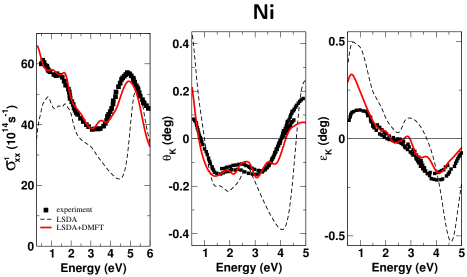

MOKE

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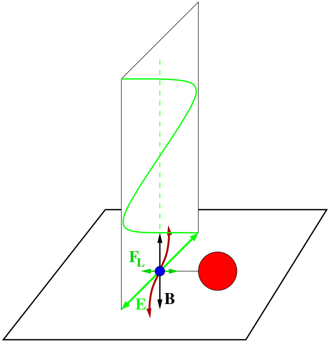
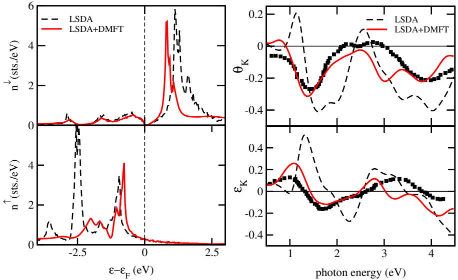
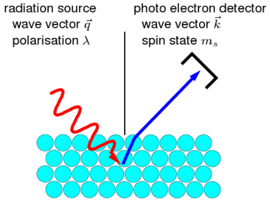


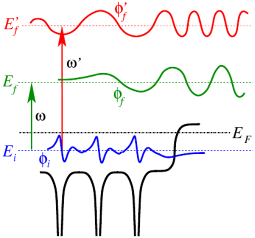
NiMnSb



S. Chadov *et al*, PRB(R) 2007

XPS





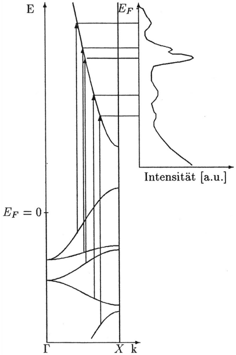
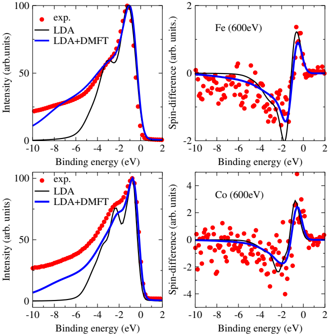


photo-current (Fermi's golden rule)

$$j \quad \propto \quad \sum_i |\langle \phi_f | \hat{\mathcal{H}}^{\vec{q}\lambda}_{rad} | \phi_i \rangle|^2 \delta(E_f - E_i - \omega)$$

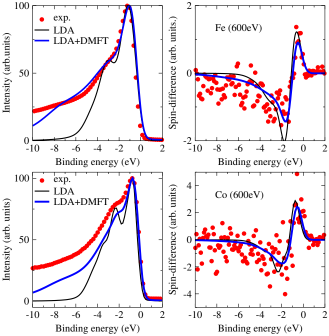
with final state $\phi_f = \mathcal{T}_R \phi^{LEED}$ — time reversed LEED state

Angle-integrated VB-XPS
in bcc-Fe and hcp-Co:



XPS

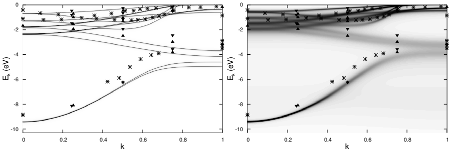
Angle-integrated VB-XPS in bcc-Fe and hcp-Co:



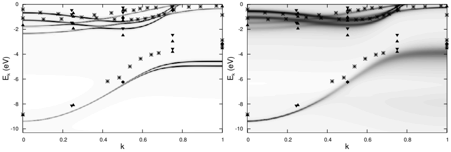
J. Minár, *et al*, PRL 2005

Angle-resolved VB-XPS for fcc-Ni:

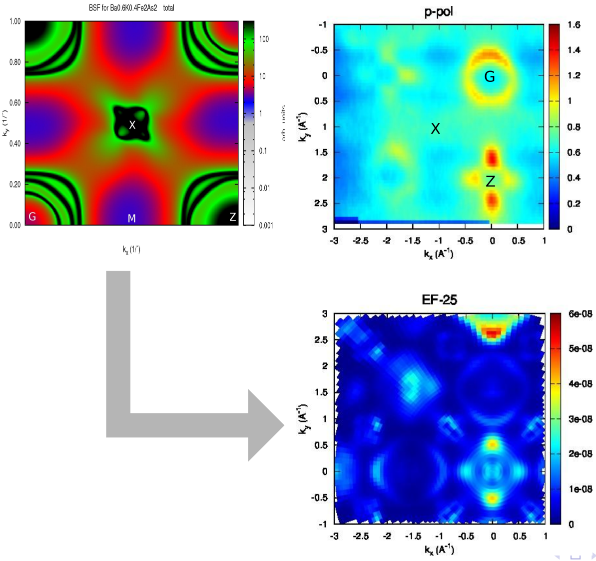
Bloch spectral functions ($\Gamma - \Delta - X$):



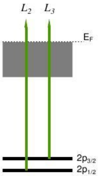
VB-XPS ($\vec{k} \parallel [001]$):

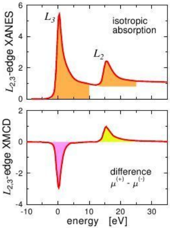


$Ba_{0.6}K_{0.4}Fe_2As_2$



XAS/XMCD



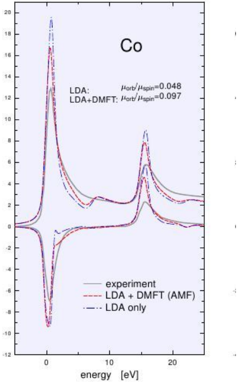


XMCD sum rules:

By adding, subtracting and dividing the peak areas, chemically-specific μ_{spin}, μ_{orb} and $\mu_{\text{orb}}/\mu_{\text{spin}}$ can be obtained

$$\int (\Delta\mu_{L_3} - 2\Delta\mu_{L_2})\, dE \quad \sim \quad \frac{\mu_{\text{spin}}^{(d)} + 7T_z^{(d)}}{3n_h^{(d)}}$$

$$\int (\Delta\mu_{L_3} + \Delta\mu_{L_2})\, dE \quad \sim \quad \frac{\mu_{\text{orb}}^{(d)}}{2n_h^{(d)}}$$



Finally

Thank you!