Orbital polarization in low transition metal systems

CENTRE NATIONAL
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SCIENTIFIQUE

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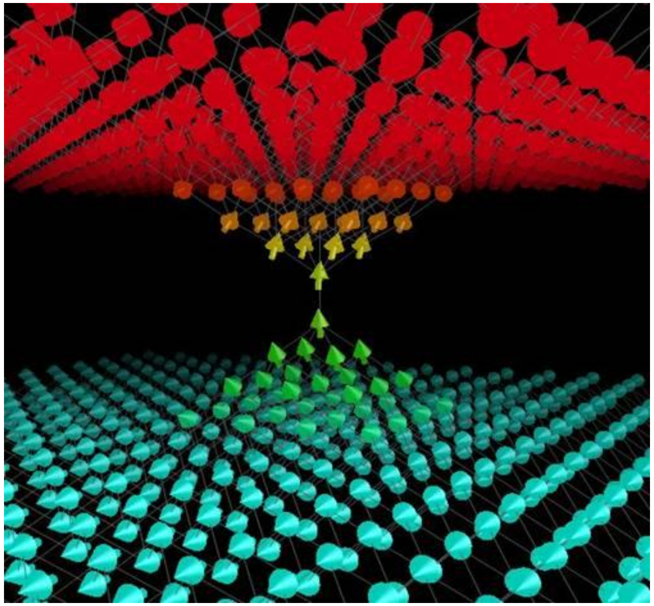


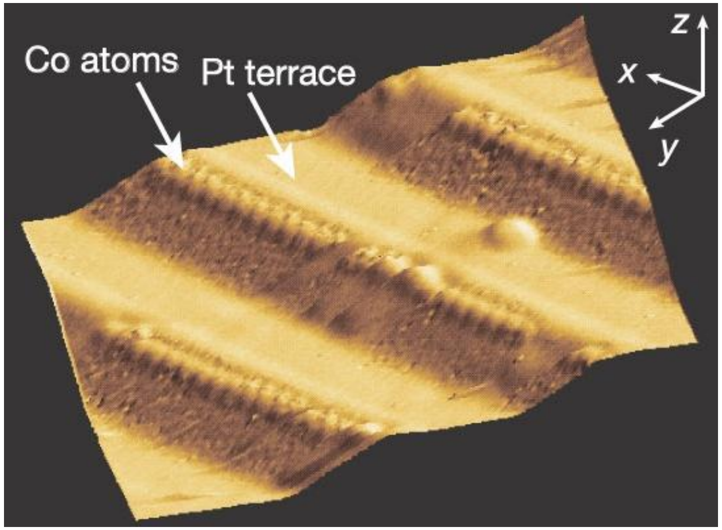
Introduction

Motivation

Quenching of orbital magnetism and MAE in bulk

In bulk (Fe, Co, Ni) $M_L \approx 0.1\mu_B$ $\text{MAE} \approx 10^{-5} eV$

Strong enhancement of orbital Magnetism and MAE in low dimension



Gambardella et al,
Nature **416**, 301 (2002)

For a monatomic wire Co/Pt(997)

$$M_L \approx 0.7\mu_B$$

$$\text{MAE} \approx 2meV$$

Introduction

What about theory

DFT

Underestimation of M_L by standard DFT calculations
(or TB Stoner like models)

$$M_L \approx 0.15\mu_B$$

$$\mathrm{MAE} \approx 1meV$$

Many versions

Possible improvements

Orbital Polarization Ansatz

$$E = E_{DFT} + \Delta E_{OP}$$

$$\Delta E_{OP} = -\tfrac{1}{2} B \langle L \rangle^2$$

$$M_L \approx 0.92\mu_B$$

LDA+U

$$E = E_{DFT} + \Delta E_{HF}$$

$$\Delta E_{HF} = E_{HF}(n_{m\sigma}) - E_{HF}(n_{m\sigma}^{LSDA})$$

$$M_L \approx 0.45\mu_B$$

Hartree Fock Hamiltonian

2-electrons intra-atomic interaction Hamiltonian

$$H_{\text{int}} = \frac{1}{2} \sum_{\lambda\mu\nu\eta} U_{\lambda\mu\nu\eta} c^{+}_{\lambda\sigma} c^{+}_{\mu\sigma'} c_{\eta\sigma'} c_{\nu\sigma}$$

Hartree Fock decoupling (HF1)

$$H^{HF1}_{\text{int}} = \frac{1}{2} \sum_{\substack{\lambda\mu\nu\eta \\ \sigma\sigma'}} \left(U_{\eta\mu\nu\lambda} \left\langle c^{+}_{\eta\sigma} c_{\nu\sigma} \right\rangle c^{+}_{\mu\sigma'} c_{\lambda\sigma'} - U_{\eta\mu\lambda\nu} \left\langle c^{+}_{\eta\sigma} c_{\nu\sigma'} \right\rangle c^{+}_{\mu\sigma'} c_{\lambda\sigma} \right)$$

Coulomb matrix elements

$$U_{\lambda\mu\nu\eta} = \left\langle \varphi_{\lambda\sigma}(\vec{r}) \varphi_{\mu\sigma'}(\vec{r}\,') \right| \frac{1}{\left| \vec{r} - \vec{r}\,' \right|} \left| \varphi_{\nu\sigma}(\vec{r}) \varphi_{\eta\sigma'}(\vec{r}\,') \right\rangle$$

Coulomb matrix elements

$$U_{\lambda\mu\nu\eta} = \text{Linear Function}(A, B, C)$$

A, B, C : Racah parameters

In cubic harmonics

2 orbitals terms

$$\left\{ \begin{array}{l} U = 1/4 \sum\limits_{\mu,\mu\neq\lambda} U_{\lambda\mu\lambda\mu} = A - B + C \\ J = 1/4 \sum\limits_{\mu,\mu\neq\lambda} U_{\lambda\mu\mu\lambda} = \frac{5}{2} B + C \end{array} \right.$$

1 orbital term

$$U_{\lambda\lambda\lambda\lambda} = U + 2J$$

3-4 orbitals terms Function of B only

New set of parameters

U, J, B

Coulomb matrix elements

In spherical harmonics

U_{mm} dependant of m

3-4 orbitals terms: function of B and C…..

Anisimov notations

$$U_A = 1/25 \sum_{m,m'} U_{mm'} = A + 7/5C$$

$$U_A - J_A = 1/20 \sum_{\substack{m,m' \\ m \neq m'}} (U_{mm'} - J_{mm'})$$

$$U_A = U + \frac{2J}{5} \qquad J_A = \frac{7}{5} J$$

$$B = 0.1 J_A$$

Simplified Hamiltonian

$B = 0$ (HF2) model

2 orbitals terms

$$\begin{cases} U_{\lambda\mu\lambda\mu} = U & \forall(\lambda,\mu), \lambda \neq \mu \\ U_{\lambda\mu\mu\lambda} = J & \forall(\lambda,\mu), \lambda \neq \mu \end{cases}$$

1 orbital terms

$$U_{\lambda\lambda\lambda\lambda} = U + 2J$$

3-4 orbitals terms

0

Stoner model (HF3)

$$H_{\text{int}}^{HF3} = \sum_{\lambda\sigma} \left(U_{eff} N - \sigma \tfrac{1}{2} IM \right) c_{\lambda\sigma}^{+} c_{\lambda\sigma}$$

Stoner parameter

$$I = (U + 6J)/5$$

Orbital Polarization ansatz

OPA energy

$$\Delta E_{OP} = -\tfrac{1}{2} B \langle L \rangle^2$$

OPA Hamiltonian

$$H_{OP} = -B \langle L_z \rangle \sum_{\lambda\mu} [L_z]_{\lambda\mu} c^+_{\lambda\sigma} c_{\mu\sigma}$$

Solovyev work

$$\text{HF1} \xrightarrow{\text{Very special case}} \text{OPA}$$

In general no obvious justification of OPA

Parameters of our model

- **TB parameters** Simplest d-band model

$$(dd\sigma, dd\pi, dd\delta) \propto (-6, 4, -1)$$

$$dd\sigma = -0.749eV \qquad 1/R^5 \quad \text{law}$$

- **Stoner parameter** $I = 0.67eV$

- **Coulomb and exchange parameters**

$$U = J = 0.48eV \qquad \text{(I=7/5U)}$$

- **Racah parameter** $B = 0.14J$

- **Spin-orbit coupling parameter** $H_{SO} = \xi L.S$

$$\xi = 0.06eV$$

Results in the bulk

Stoner parameter: **chosen to reproduce the spin-moment**

HF2 $M_S = 2.12\mu_B$ $M_L = 0.08\mu_B$

HF1 $M_S = 2.11\mu_B$ $M_L = 0.12\mu_B$

Slight increase of the orbital moment

Monatomic wire

$d = 4.7 a.u.$

saturated

	HF1	HF2	HF2	HF3	HF3
			OPA		OPA
Ms ∥	3	3	3	3	3
Ms⊥	3	3	3	3	3
Lz ∥	1.45	0.37	1.31	0.37	1.31
Lz ⊥	0.49	0.25	0.61	0.25	0.60
MAE	23.4	0.7	22.3	0.6	22.3

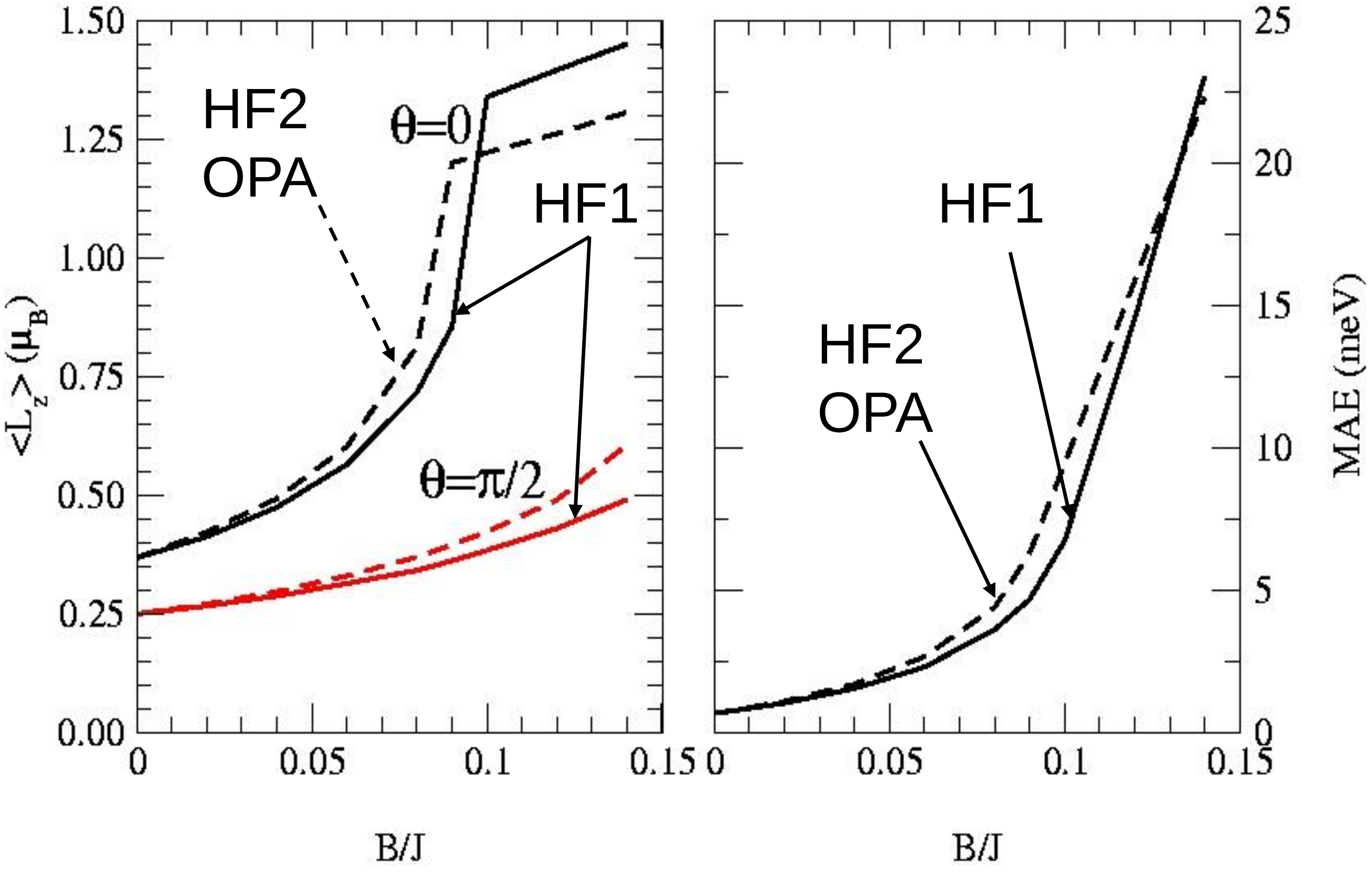
$d = 4.25 a.u.$

unsaturated

	HF1	HF2	HF2	HF3	HF3
			OPA		OPA
Ms ∥	1.51	1.24	1.23	0.94	0.78
Ms⊥	1.51	1.23	1.24	0.93	0.94
Lz ∥	0.33	0.19	0.39	0.24	1.07
Lz ⊥	0.21	0.10	0.18	0.08	0.15
MAE	-0.7	-0.3	1.5	0.0	6.2

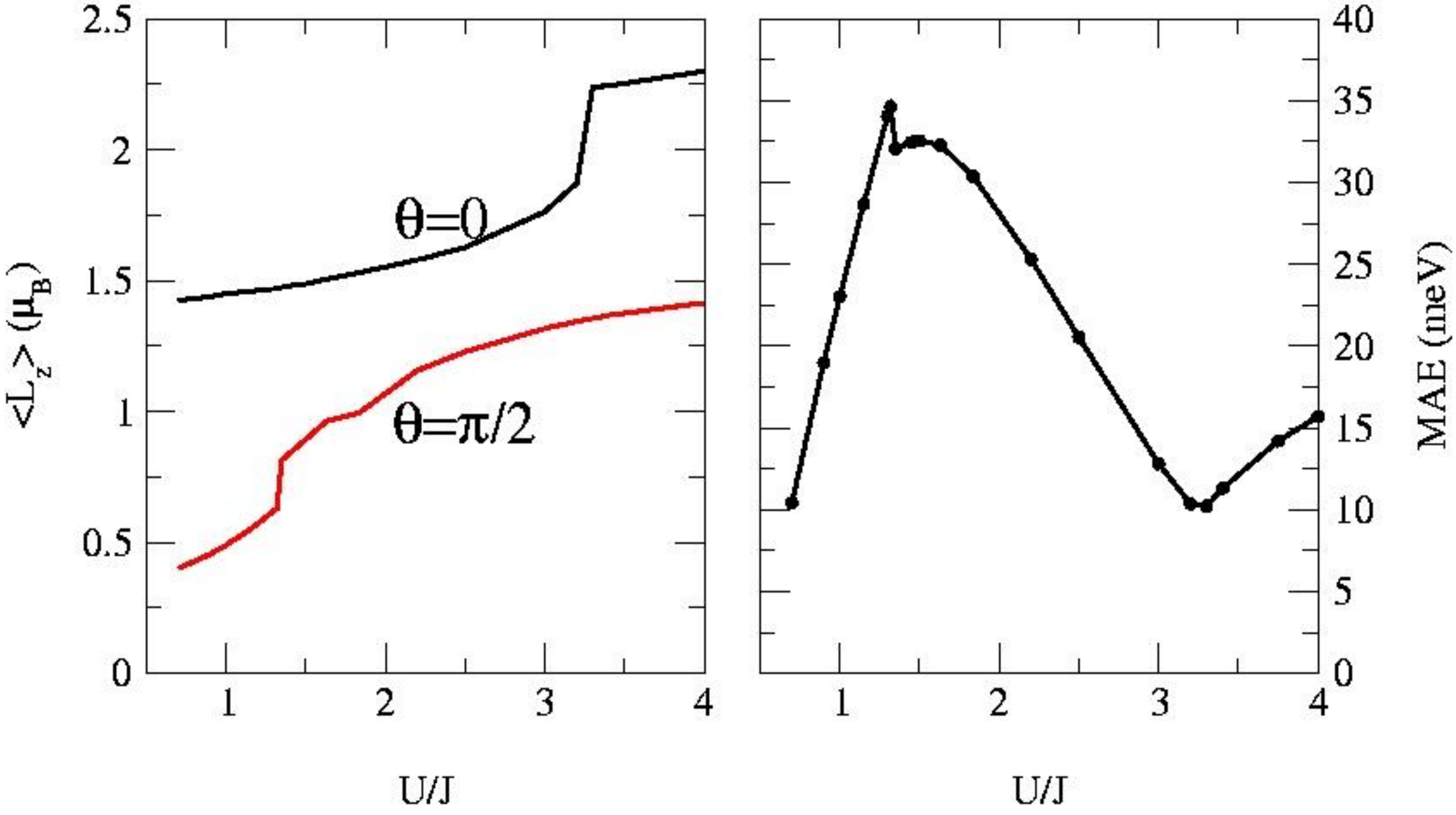
Monatomic wire

Varying the parameters (B/J)



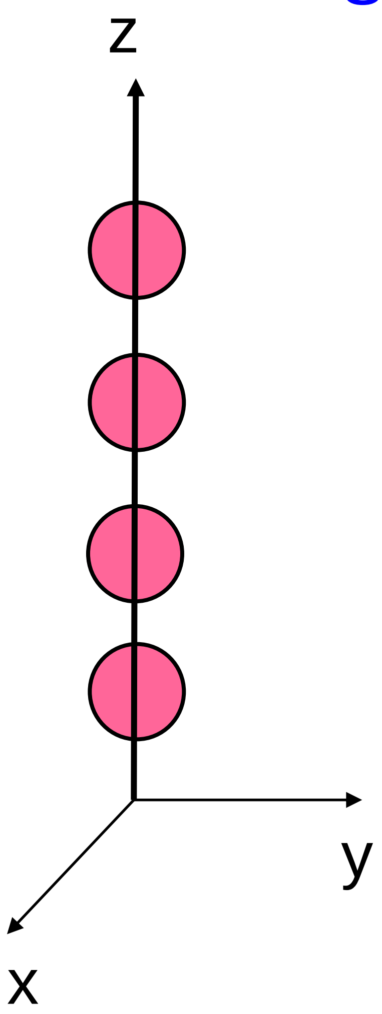
Monatomic wire

Varying the parameters (U/J), I constant

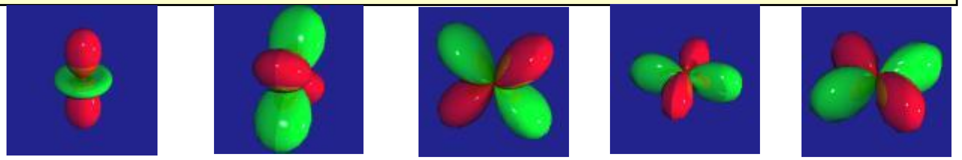


Monatomic wire

Slater Koster matrix of the d-band linear chain

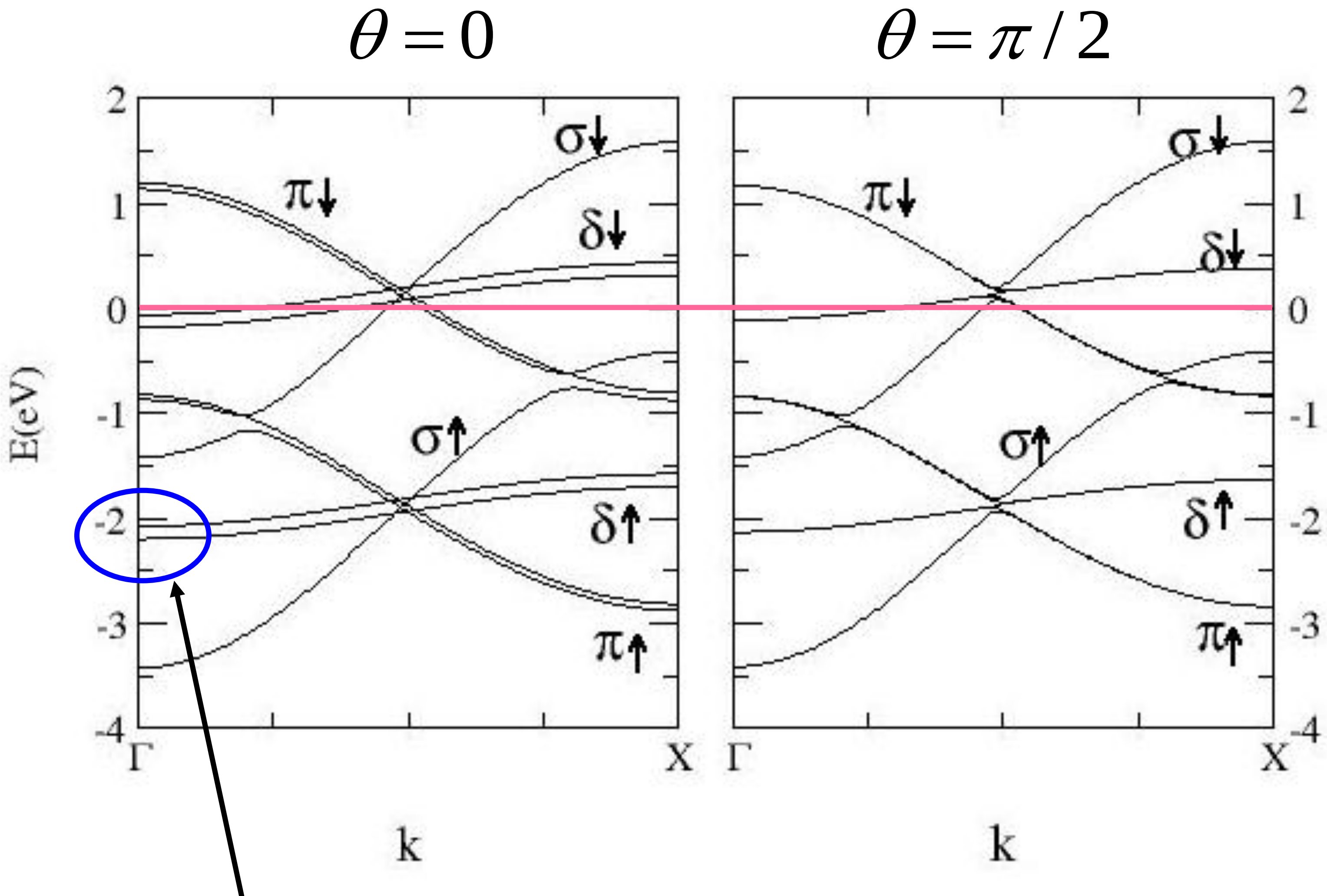


$$\begin{array}{cccccc}
 & d_{3z^2-r^2} & d_{zx} & d_{yz} & d_{xy} & d_{x^2-y^2} \\
d_{3z^2-r^2} & dd\sigma & 0 & 0 & 0 & 0 \\
d_{zx} & 0 & dd\pi & 0 & 0 & 0 \\
d_{yz} & 0 & 0 & dd\pi & 0 & 0 \\
d_{xy} & 0 & 0 & 0 & dd\delta & 0 \\
d_{x^2-y^2} & 0 & 0 & 0 & 0 & dd\delta
\end{array}$$



Monatomic wire

HF3



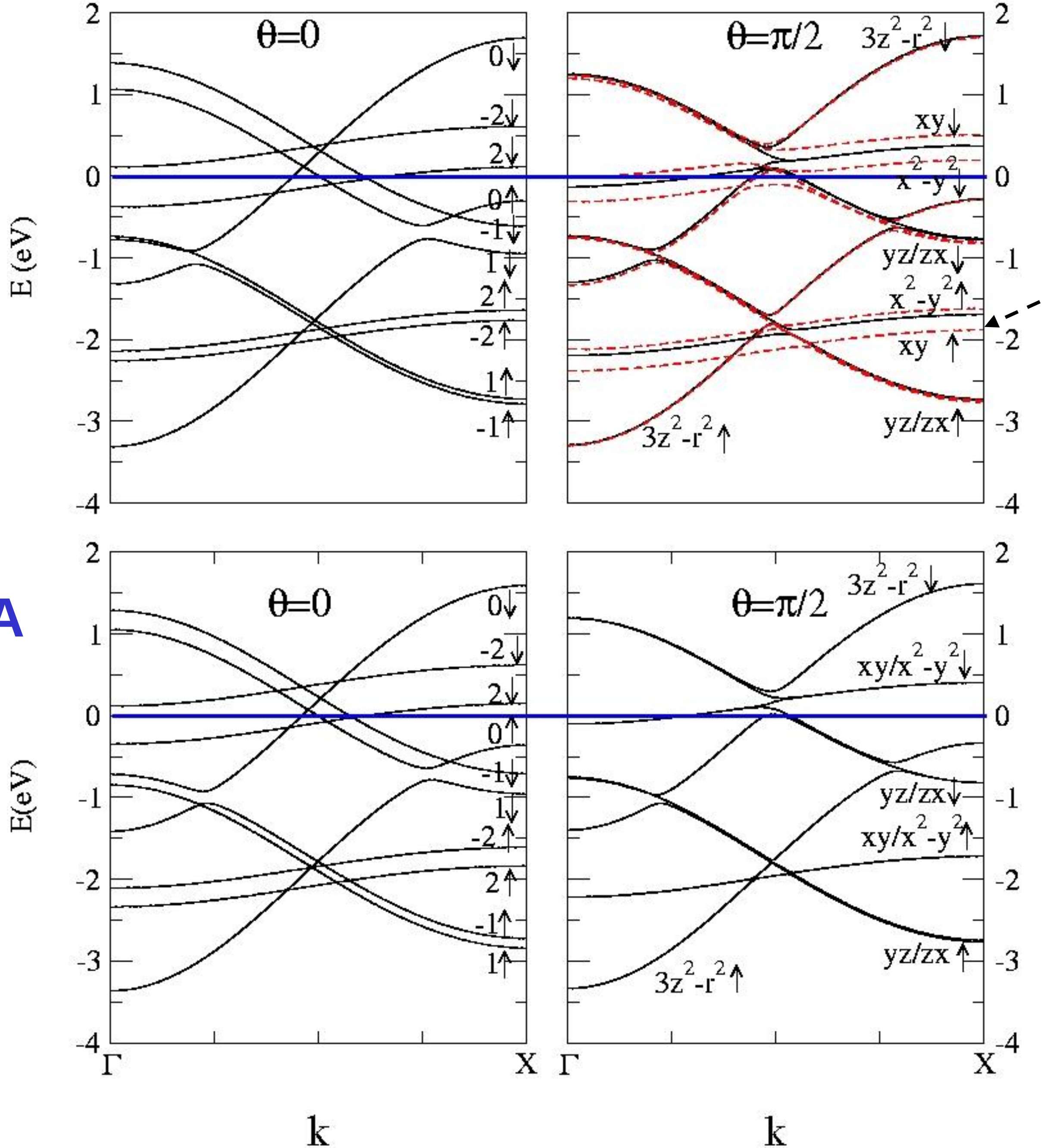
splitting 2ξ

No splitting but band avoiding

Monatomic wire

HF1

HF2+OPA



$U / J = 1.34$

CONCLUSION

- Large orbital moment and MAE in low dimension.
- HF3+OPA is not accurate enough for unsaturated systems.
- HF1 is necessary in low dimensional systems.
- Giant magnetoresistance in low dimensional systems

PERSPECTIVES

- Extend our model to more realistic Hamiltonians
  - spd TB (almost done)
  - generalized L(S)DA+U (J,B).
- Study of more complex nanostructures
- Influence on transport properties (magnetoresistance)
- Determination of physically acceptable U,J,B!!!