Magnetism and SOC in Wien2k

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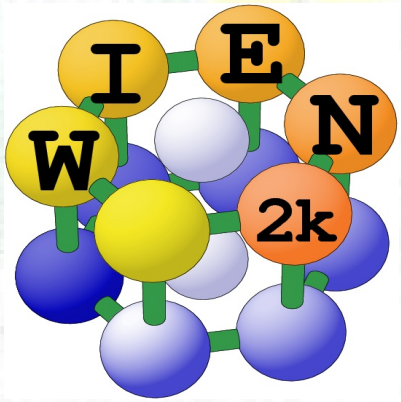


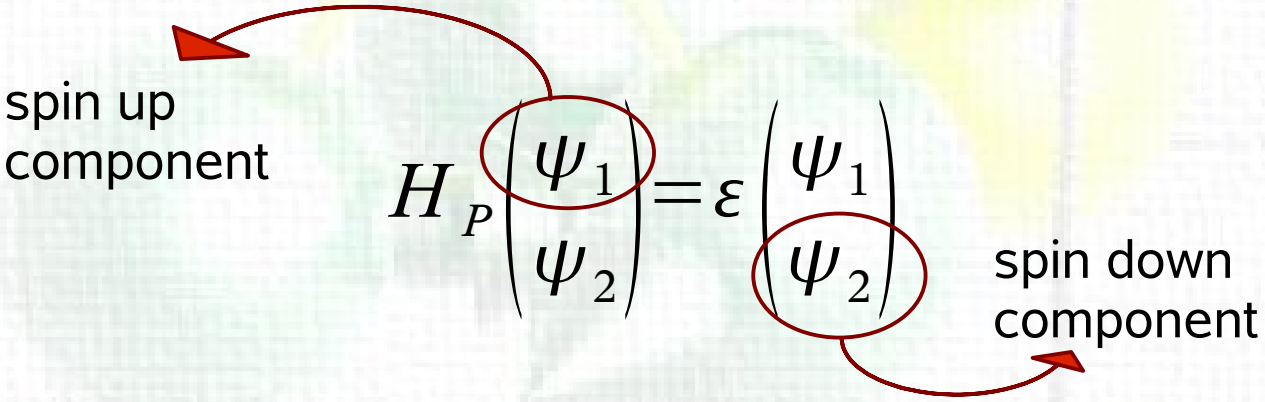
SCOPE

- magnetism in Wien2k
  - collinear spins (ferro, ferri, antiferro-magnets)
  - non-collinear spin (any arrangements), introduction to WienNCM
- spin-orbit coupling (SOC) in Wien2k

Pauli Hamiltonian

$$H_P = -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \mu_B \vec{\sigma}\cdot\vec{B}_{ef} + \zeta(\vec{\sigma}\cdot\vec{l}) \ldots$$

- 2x2 matrix in spin space, due to Pauli spin operators
- wave function is a 2-component vector (spinor)



Pauli Hamiltonian

$$H_P = -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \mu_B \vec{\sigma}\cdot\vec{B}_{ef} + \zeta(\vec{\sigma}\cdot\vec{l})\ldots$$

electrostatic potential

magnetic field

spin-orbit coup.

$$V_{ef} = V_{ext} + V_H + V_{xc}$$

Hartree term

exchange-correlation potential

$$B_{ef} = B_{ext} + B_{xc}$$

exchange-correlation field

- exchange-correlation potential V_{xc} and magnetic field B_{xc} are defined within DFT LDA or GGA

Exchange and correlation

- from DFT LDA exchange-correlation energy:

$$E_{xc}(n,\vec{m}) = \int n\,\epsilon_{xc}(n,\vec{m})\,dr^3$$

local function of n and m

- definition of V_{cx} and B_{xc}:

$$V_{xc} = \frac{\partial E_{xc}(n,\vec{m})}{\partial n} \qquad \vec{B}_{xc} = \frac{\partial E_{xc}(n,\vec{m})}{\partial \vec{m}}$$

functional derivatives

- LDA expression for V_{cx} and B_{xc}:

$$V_{xc} = \epsilon_{xc}(n,\vec{m}) + n\frac{\partial \epsilon_{xc}(n,\vec{m})}{\partial n}$$

B_{xc} and m are parallel

$$\vec{B}_{xc} = n\frac{\partial \epsilon_{xc}(n,\vec{m})}{\partial m}\hat{m}$$

Non-collinear case

$$H_P = -\frac{\hbar}{2\mathrm{m}}\nabla^2 + V_{ef} + \mu_B \vec{\sigma}\cdot\vec{B}_{ef} + \zeta(\vec{\sigma}\cdot\vec{l})\ \ldots$$

- direction of magnetization vary in space
- spin-orbit coupling is present

$$\begin{pmatrix} -\frac{\hbar}{2\mathrm{m}}\nabla^2 + V_{ef} + \mu_B B_z + \ldots & \mu_B\left(B_x - iB_y\right) \\ \mu_B\left(B_x + iB_y\right) & -\frac{\hbar}{2\mathrm{m}}\nabla^2 + V_{ef} + \mu_B B_z + \ldots \end{pmatrix} \psi = \varepsilon\,\psi$$

$$\psi = \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix}, \quad \psi_1, \psi_2 \neq 0$$

- solutions are non-pure spinors
- non-collinear magnetic moments

Collinear case

$$H_P = -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \mu_B \vec{\sigma}\cdot\vec{B}_{ef} + \cancel{\zeta(\vec{\sigma}\cdot\vec{l})} \ldots$$

- magnetization in Z direction, B_x and B_y=0
- spin-orbit coupling is not present

$$\begin{pmatrix} -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \mu_B B_z + \ldots & 0 \\ 0 & -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \mu_B B_z + \ldots \end{pmatrix} \psi = \varepsilon\,\psi$$

$$\psi_\uparrow = \begin{pmatrix} \psi_1 \\ 0 \end{pmatrix},\ \psi_\downarrow = \begin{pmatrix} 0 \\ \psi_2 \end{pmatrix},\quad \varepsilon_\uparrow \neq \varepsilon_\downarrow$$

- solutions are pure spinors
- collinear magnetic moments

Non-magnetic case

$$H_P = -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \cancel{\mu_B \vec{\sigma}\cdot\vec{B}_{ef}} + \cancel{\zeta(\vec{\sigma}\cdot\vec{l})} \ldots$$

- no magnetization present, B_x, B_y and B_z=0
- spin-orbit coupling is not present

$$\begin{pmatrix} -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \ldots & 0 \\ 0 & -\frac{\hbar}{2m}\nabla^2 + V_{ef} + \ldots \end{pmatrix} \psi = \varepsilon\,\psi$$

$$\psi_\uparrow = \begin{pmatrix} \psi \\ 0 \end{pmatrix},\ \psi_\downarrow = \begin{pmatrix} 0 \\ \psi \end{pmatrix},\ \ \varepsilon_\uparrow = \varepsilon_\downarrow$$

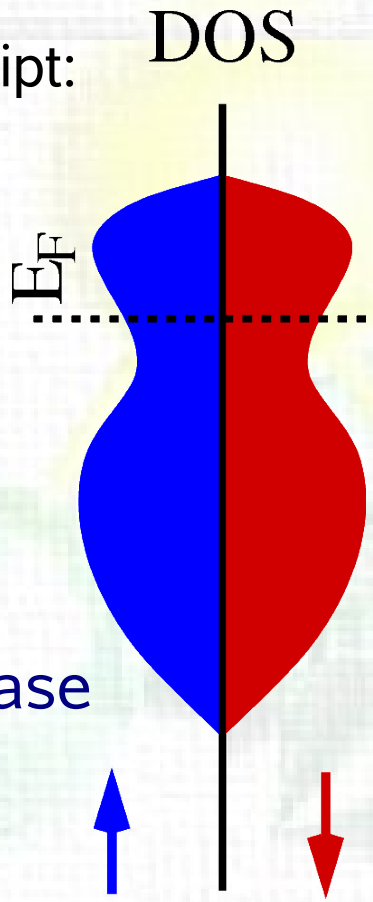
- solutions are pure spinors
- degenerate spin solutions

Magnetism and Wien2k

- Wien2k can only handle collinear or non-magnetic cases

run_lapw script:

```
x lapw0
x lapw1
x lapw2
x lcore
x mixer
```



non-magnetic case

$$m = n_\uparrow - n_\downarrow = 0$$

Magnetism and Wien2k

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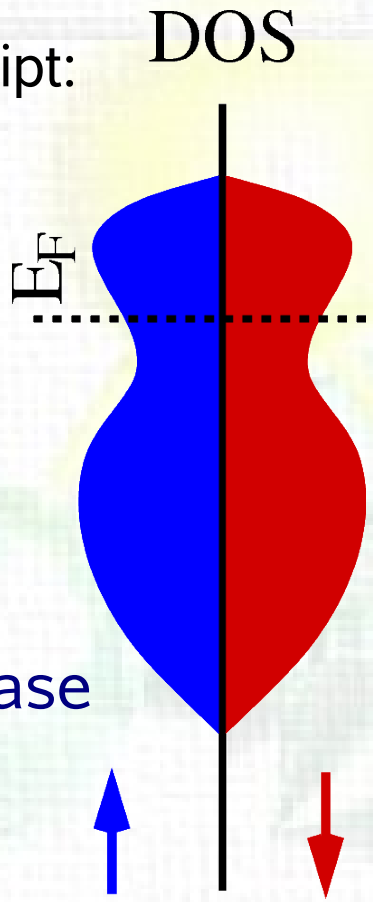
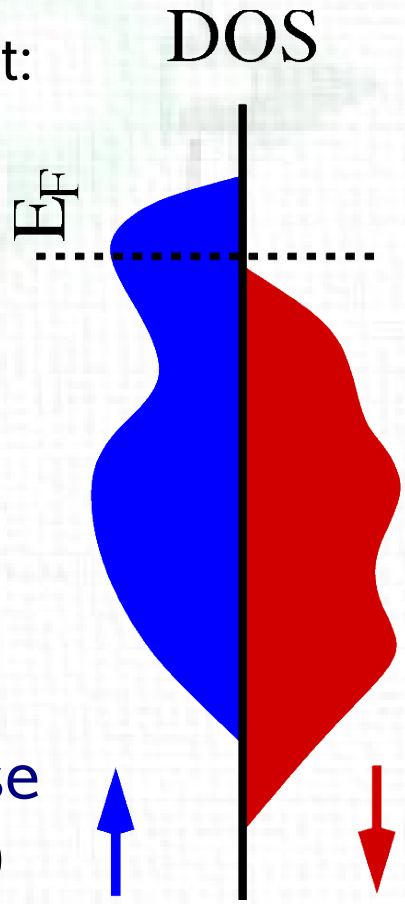


non-magnetic case

$$m = n_\uparrow - n_\downarrow = 0$$

run_lapw script:

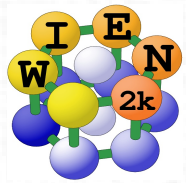
```
x lapw0
x lapw1 -up
x lapw1 -dn
x lapw2 -up
x lapw2 -dn
x lcore -up
x lcore -dn
x mixer
```



magnetic case

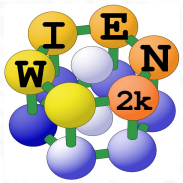
$$m = n_\uparrow - n_\downarrow \neq 0$$

Spin polarized calculations



- *runsp_lapw* script (unconstrained magnetic calc.)
  - runs lapw1/2 for both spins independently
  - case.scf contains extra information:
    - `grep :MMT case.scf` (for total moment)
    - `grep :MMI case.scf` (for atomic moments)
    - `grep :HFF case.scf` (for hyperfine fields)

Spin polarized calculations



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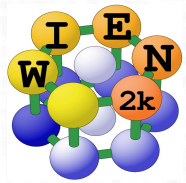
    - `grep :HFF case.scf` (for hyperfine fields)
- *runfsm_lapw -m value* (constrained moment calc.)
  - for difficult to converge magnetic cases or simply to constrain a moment (→ 2 Fermi-energies → external magnetic field)
- *runafm_lapw* (anti-ferromagnetic calculation)
  - calculates only spin-up, uses symmetry to generate spin-dn

Spin polarized calculations



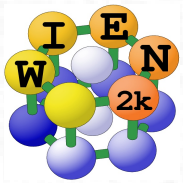
- *runsp_lapw* script (unconstrained magnetic calc.)
- *runfsm_lapw -m value* (constrained moment calc.)
- *runafm_lapw* (anti-ferromagnetic calculation)

  - spin-orbit coupling can be included in second variational step
  - never mix polarized and non-polarized calculations in one *case* directory !!!

Non-collinear calculations

- in a case of non-collinear spin arrangements WienNCM (Wien2k clone) has to be used
  - code based on Wien2k (available for Wien2k users)
  - structure and usage philosophy similar to Wien2k
  - independent source tree, independent installation

Non-collinear calculations



- case of non-collinear spin arrangements WienNCM (Wien2k clone) has to be used
  - code based on Wien2k (available for Wien2k users)
  - structure and usage philosophy similar to Wien2k
  - independent source tree, independent installation
- WienNCM properties:
  - real and spin symmetry (simplifies SCF, less k-points)
  - constrained or unconstrained calculations (optimizes magnetic moments)
  - SOC in first variational step, LDA+U
  - spin spirals

WienNCM - implementation

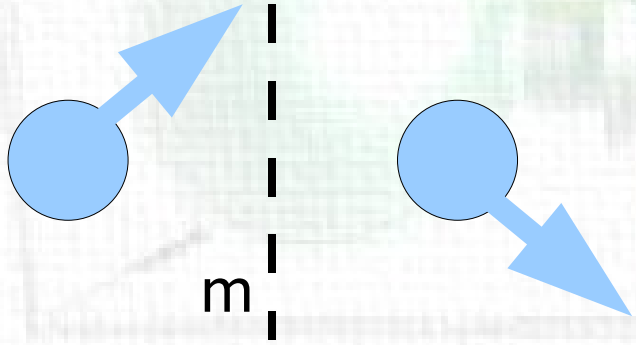
- basis set – mixed spinors (Yamagami, PRB (2000); Kurtz PRB (2001)

interstitials: $\varphi_{\vec{G}\sigma} = e^{i(\vec{G}+\vec{k})\cdot\vec{r}}\chi_\sigma$ $\qquad \chi_\sigma = \begin{pmatrix}1\\0\end{pmatrix}, \begin{pmatrix}0\\1\end{pmatrix}$

spheres: $$\varphi^{APW}_{\vec{G}\sigma} = \sum_{\sigma_\alpha}\sum_{lm}\left(A^{\vec{G}\sigma\sigma_\alpha}_{lm}u^{\sigma_\alpha}_l + B^{\vec{G}\sigma\sigma_\alpha}_{lm}\dot{u}^{\sigma_\alpha}_l\right)Y_{lm}\chi_{\sigma_\alpha}$$

$$\varphi^{APW}_{\vec{G}\sigma_\alpha} = \left(A^{\vec{G}\sigma\sigma_\alpha}_{lm}u^{\sigma_\alpha}_l + B^{\vec{G}\sigma\sigma_\alpha}_{lm}\dot{u}^{\sigma_\alpha}_l + C^{\vec{G}\sigma\sigma_\alpha}_{lm}u^{\sigma_\alpha}_{2,l}\right)Y_{lm}\chi_{\sigma_\alpha}$$

- real and spin space parts of symmetry op. are bounded



  - symmetry treatment like SOC always on
  - tool for setting up magnetic configuration
  - concept of magnetic and non-magnetic atoms

WienNCM implementation

- sphere Hamiltonian: $\hat{H} = -\frac{\hbar}{2\mathrm{m}}\nabla^2 + \hat{V} + \hat{H}_{so} + \hat{H}_{orb} + \hat{H}_c$

AMA and full NC calculation

$$\hat{V}_{FULL} = \begin{pmatrix} V_{\uparrow\uparrow} & V_{\downarrow\uparrow} \\ V_{\uparrow\downarrow} & V_{\downarrow\downarrow} \end{pmatrix} \qquad \hat{V}_{AMA} = \begin{pmatrix} V_{\uparrow\uparrow} & 0 \\ 0 & V_{\downarrow\downarrow} \end{pmatrix}$$

SOC in first diagonalization

$$\hat{H}_{so} = \xi\,\vec{\sigma}\cdot\vec{l} = \xi \begin{pmatrix} \hat{l}_z & \hat{l}_x - i\,\hat{l}_y \\ \hat{l}_x + i\,\hat{l}_y & -\hat{l}_z \end{pmatrix}$$

diagonal orbital field

$$\hat{H}_{orb} = \sum_{m\,m'} \begin{pmatrix} |m\rangle V^{\uparrow}_{mm'} \langle m'| & 0 \\ 0 & |m\rangle V^{\downarrow}_{mm'} \langle m'| \end{pmatrix}$$

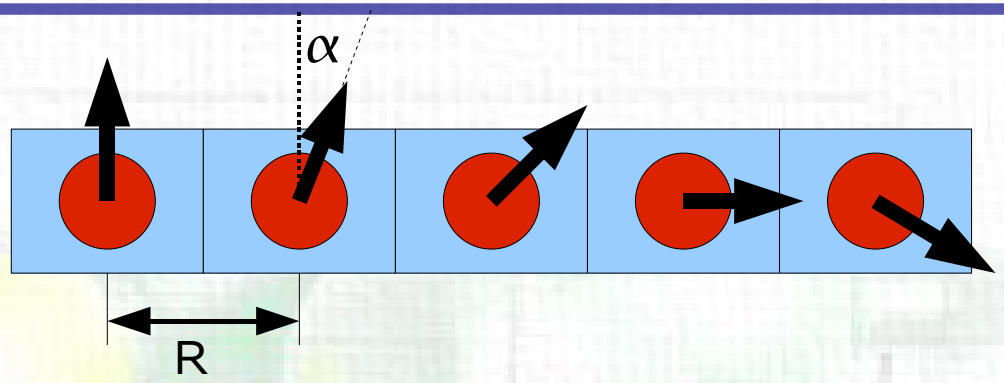
constraining field

$$\hat{H}_c = \mu_B\,\vec{\sigma}\cdot\vec{B}_c = \begin{pmatrix} 0 & \mu_B\left(B_{cx} - iB_{cy}\right) \\ \mu_B\left(B_{cx} + iB_{cy}\right) & 0 \end{pmatrix}$$

WienNCM – spin spirals

- transverse spin wave

$$\alpha = \vec{R} \cdot \vec{q}$$



- generalized Bloch theorem
  - generalized translations $T_n = \left\{ -\vec{q} \cdot \vec{R}_n \middle| \epsilon \middle| \vec{R}_n \right\}$
  - group of T_n is Abelian

$$T_n \psi_{\vec{k}}(\vec{r}) = U\left(-\vec{q} \cdot \vec{R}\right) \psi_{\vec{k}}\left(\vec{r} + \vec{R}\right) = \psi_{\vec{k}}(\vec{r})$$

$$\psi_{\vec{k}}(\vec{r}) = \mathrm{e}^{i\left(\vec{k} \cdot \vec{r}\right)} \begin{pmatrix} \mathrm{e}^{\frac{i\vec{q} \cdot \vec{r}}{2}} u^{\uparrow}(\vec{r}) \\ \mathrm{e}^{\frac{-i\vec{q} \cdot \vec{r}}{2}} u^{\downarrow}(\vec{r}) \end{pmatrix}$$

- efficient way for calculation of spin waves, only one unit cell is necessary for even incommensurate wave

WienNCM – case.inncm file

- `case.inncm` – magnetic structure file

```
FULL
  0.000000   0.000000  0.000000
 45.00000   54.73561    0
135.00000  125.26439    0
-135.00000  54.73561    0
 -45.00000 125.26439    0
 45.00000   54.73561    0
 45.00000   54.73561    0
315.00000  125.26439    0
315.00000  125.26439    0
135.00000  125.26439    0
135.00000  125.26439    0
225.00000   54.73561    0
225.00000   54.73561    0
  0.50000
```

q spiral vector

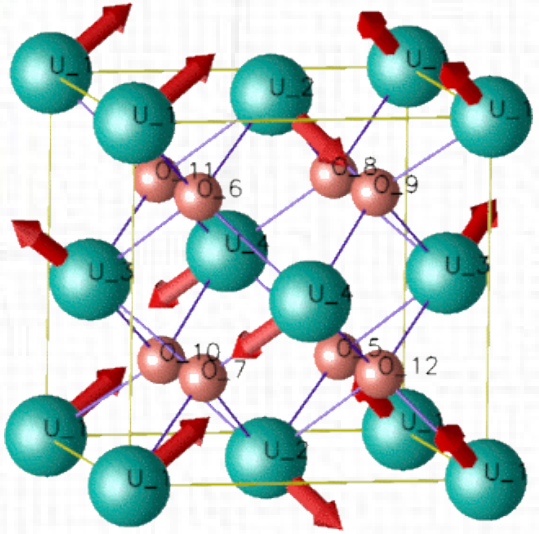
polar angles of mm

optimization switch

U, magnetic atoms

O, non-magnetic atoms

mixing for constraining field



SOC in Wien2k

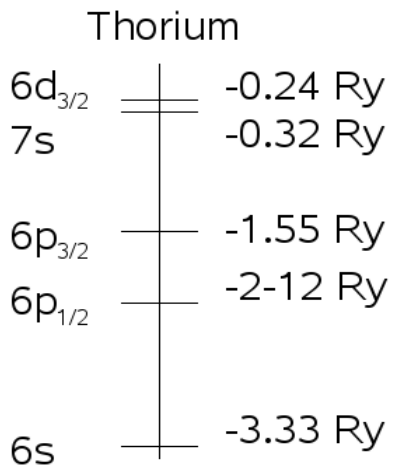
SOC in Wien2k

- Non-relativistic limit of Dirac equation

$$\left[\underbrace{\frac{p^2}{2m}+V}_{\text{Schrödinger Equation}}\underbrace{-\frac{p^4}{8m^3c^2}-\frac{\hbar^2}{4m^2c^2}\frac{dV}{dr}\frac{\partial}{\partial\vec{r}}}_{\text{mass enhancement + Darwin term}}+\underbrace{\frac{1}{2m^2c^2}\frac{1}{r}\frac{dV}{dr}(\vec{l}\,\vec{s})}_{\text{spin-orbit coupling}}\right]\Phi=\varepsilon\Phi$$

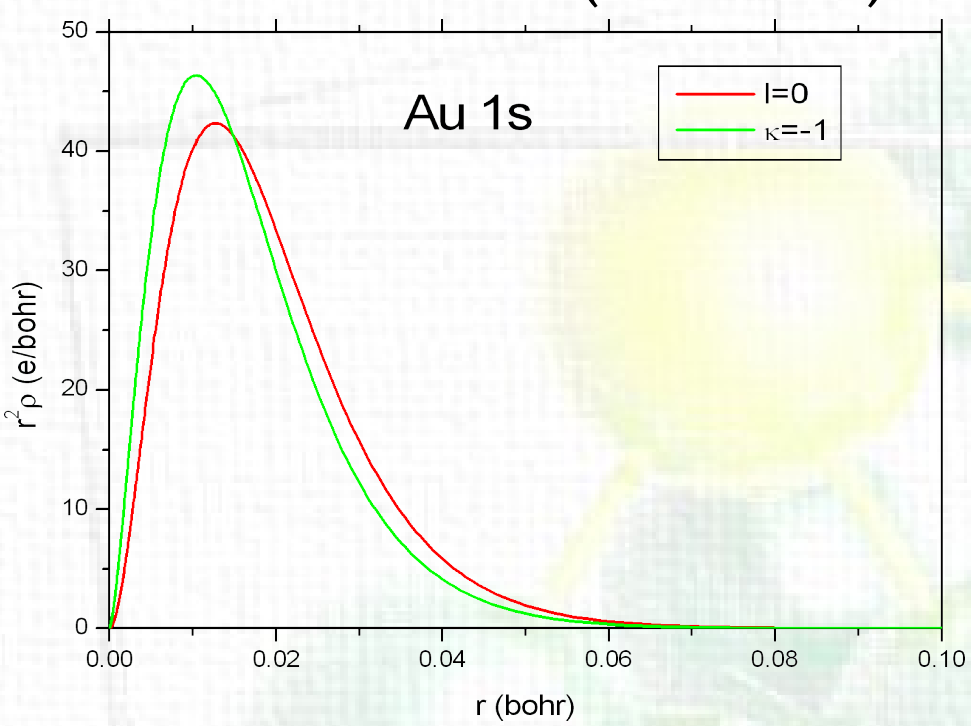
- SOC mixes up and down states, *j=l+s* is good quantum number

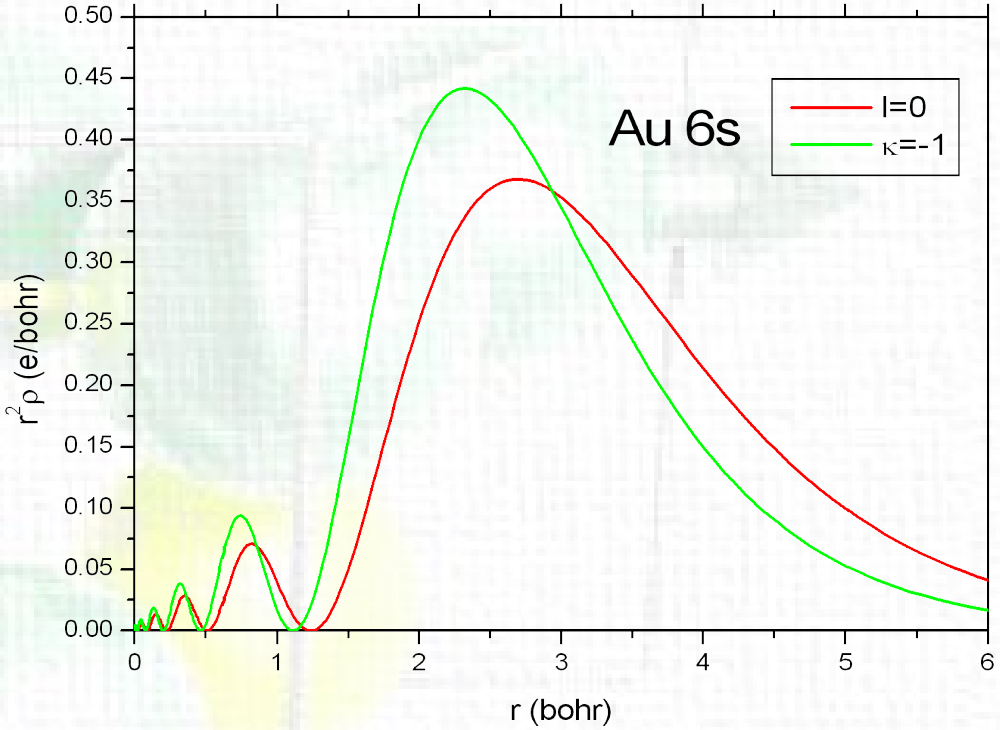
		j=l+s/2		κ=-s(j+½)		occupation	
	l	s=-1	s=+1	s=-1	s=+1	s=-1	s=+1
s	0		1/2		-1		2
p	1	1/2	3/2	1	-2	2	4
d	2	3/2	5/2	2	-3	4	6
f	3	5/2	7/2	3	-4	6	8



Relativistic orbital contraction

- Au s orbitals (no SOC)



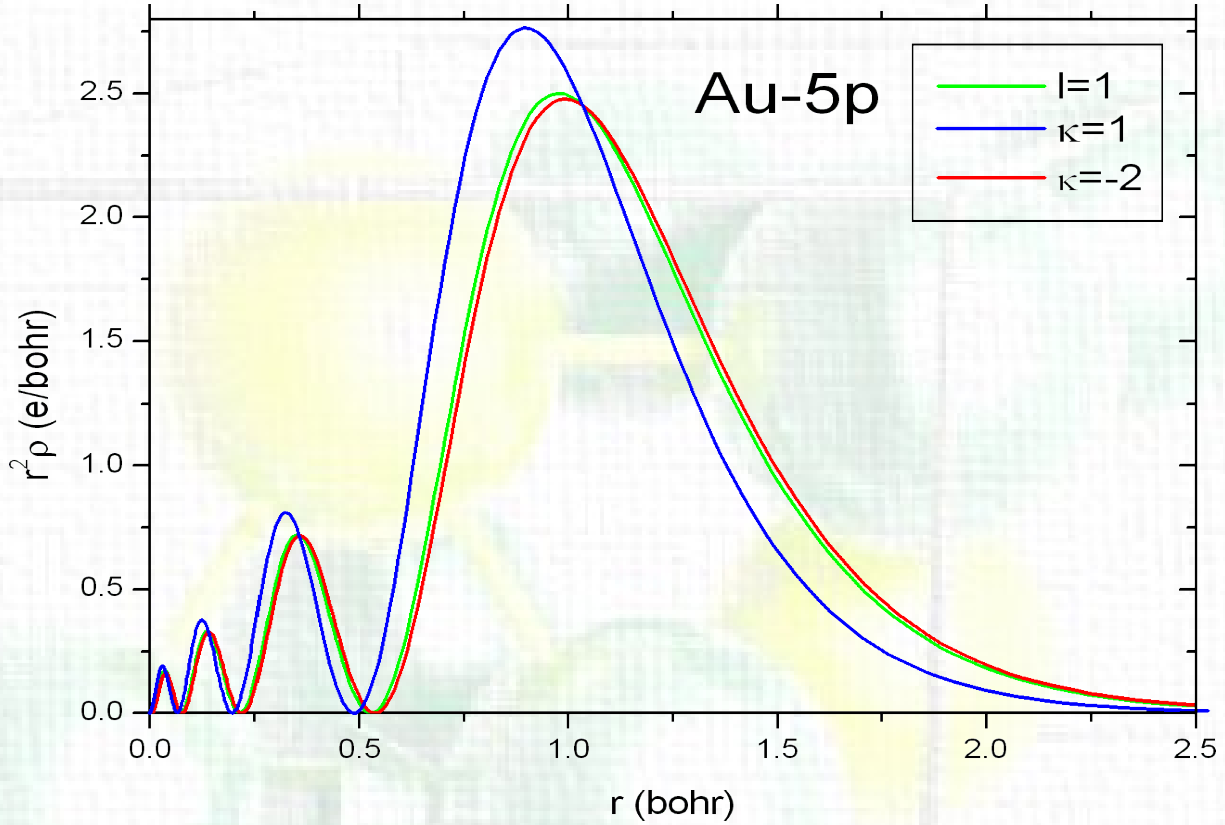


- 1s contracts due to relativistic mass enhancement
- 2s - 6s contract due to orthogonality to 1s

$$M = m / \sqrt{1 - (v/c)^2}$$

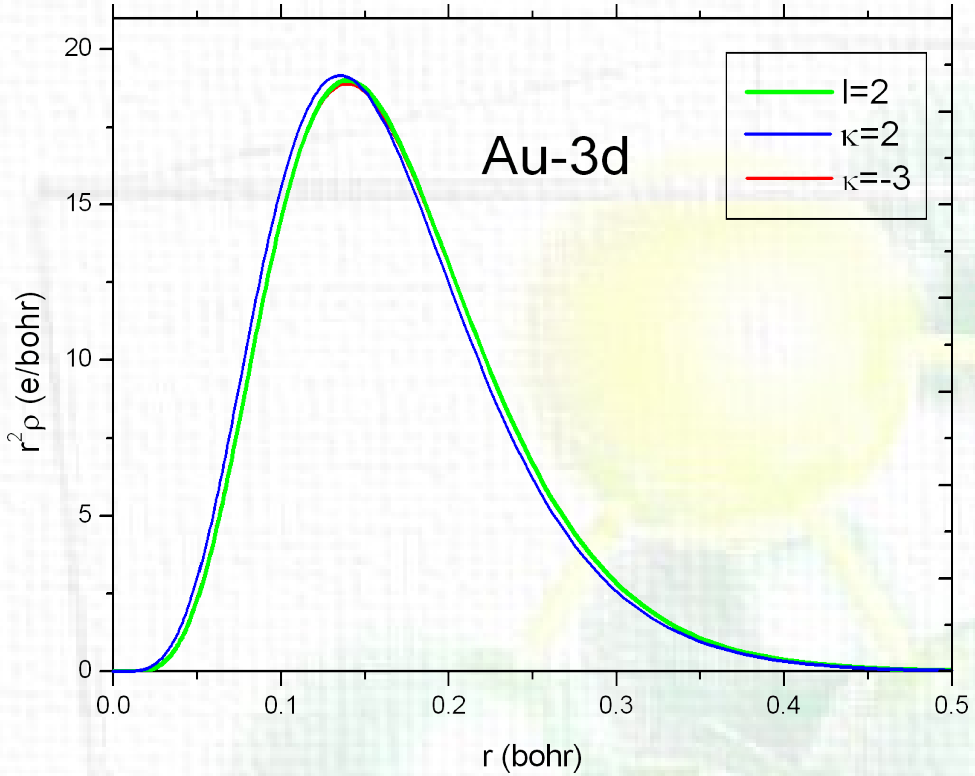
v proportional Z: Gold: Z = 79;M = 1.2 m

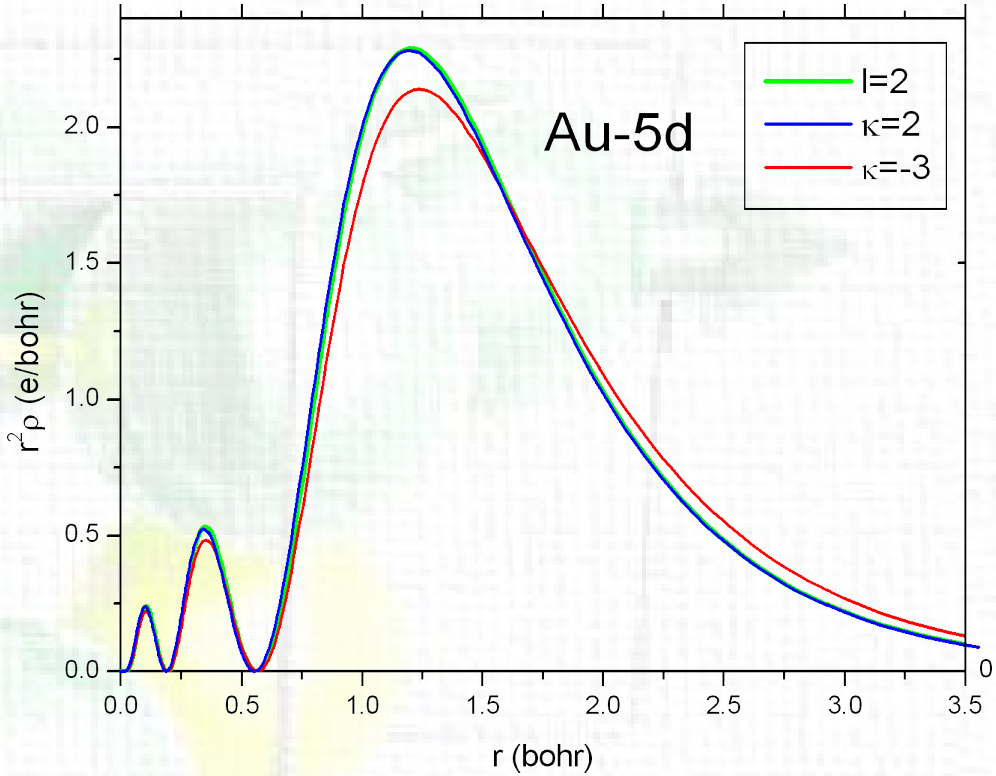
SOC splitting of p states



- Spin Orbit splitting of l-quantum number.
- $p_{1/2}$ (κ=1): markedly different behavior than non-relativistic p-state
- $u_{\kappa=1}$: non-zero at nucleus

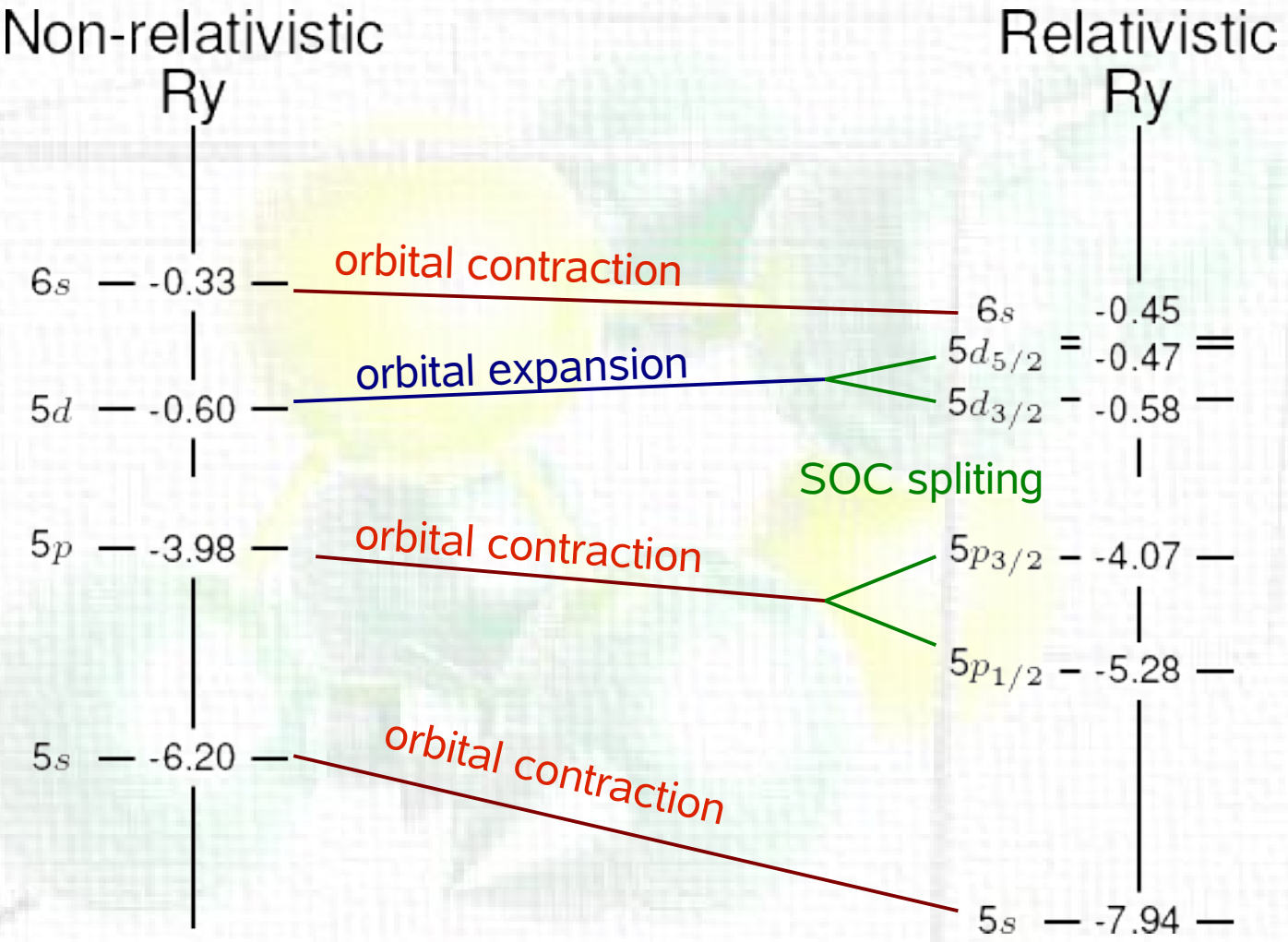
Relativistic orbital expansion





- Higher l-quantum number states expand due to better shielding of core charge from contracted s-states.

Au atomic spectra

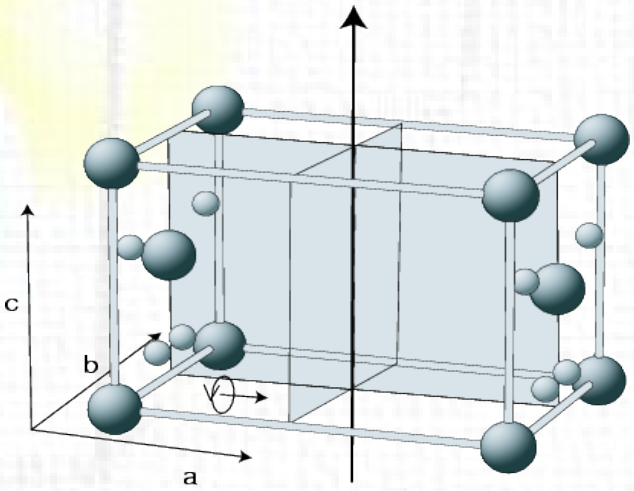


SOC in magnetic systems

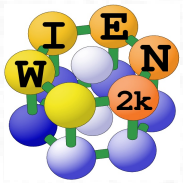
- SOC couples spin to the lattice (magneto- crystalline anisotropy)
  - direction of the exchange field matters (input in `case.inso`)
- symmetry operations acts in real and spin space
  - number of symmetry operations may be reduced
  - time inversion is not symmetry operation (no add inversion for k-list)
  - initso_lapw (symmetso) detects new symmetry setting

direction of magnetization

	[100]	[010]	[001]	[110]
1	A	A	A	A
m_a	A	B	B	-
m_b	B	A	B	-
2_z	B	B	A	B

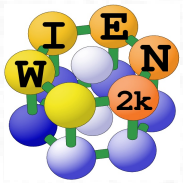


SOC in Wien2k



- WIEN2k offers several levels of treating relativity:
  - non-relativistic: select NREL in case.struct (not recommended)
  - standard: fully-relativistic core, scalar-relativistic valence
    - mass-velocity and Darwin s-shift, no spin-orbit interaction
  - "fully"-relativistic:
    - adding SO in "second variation" (using previous eigenstates as basis)
    - adding p-1/2 LOs to increase accuracy (caution!!!)
    - Non-magnetic systems:
      - SO does NOT reduce symmetry. initso_lapw just generates case.inso and case.in2c.
    - Magnetic systems:
      - symmetso detects proper symmetry and rewrites case.struct/in*/clm*

SOC in Wien2k

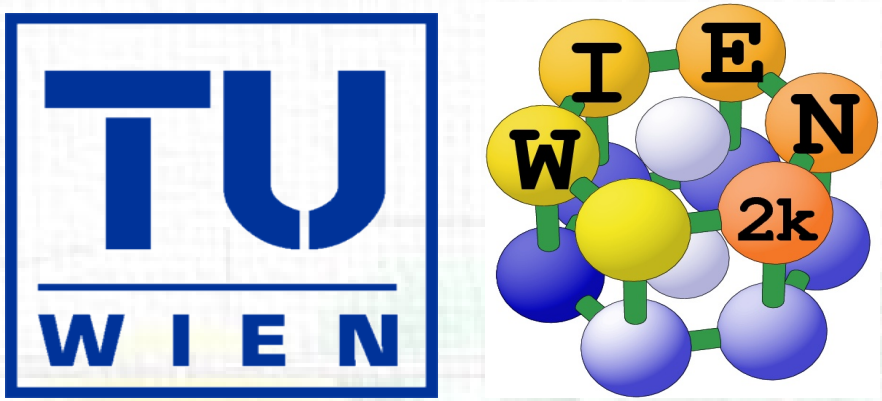


- run(sp)_lapw -so script:

```
x lapw1          (increase E-max for more eigenvectors in second diag.)
x lapwso         (second diagonalization)
x lapw2 -so -c   (SO ALWAYS needs complex lapw2 version)
```

- `case.inso` file:

```
WFFIL
4  1  0                    llmax,ipr,kpot
-10.0000   1.50000         emin,emax (output energy window)
 0.  0.  1.                direction of magnetization (lattice vectors)
 1                         number of atoms for which RLO is added
 2   -0.97     0.005       atom number,e-lo,de (case.in1), repeat NX times
 0 0 0 0 0                 number of atoms for which SO is switched off; atoms
```



Thank you for your attention