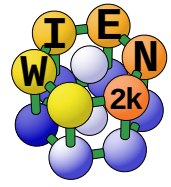
Non-collinear magnetism in WIEN2k



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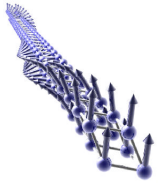
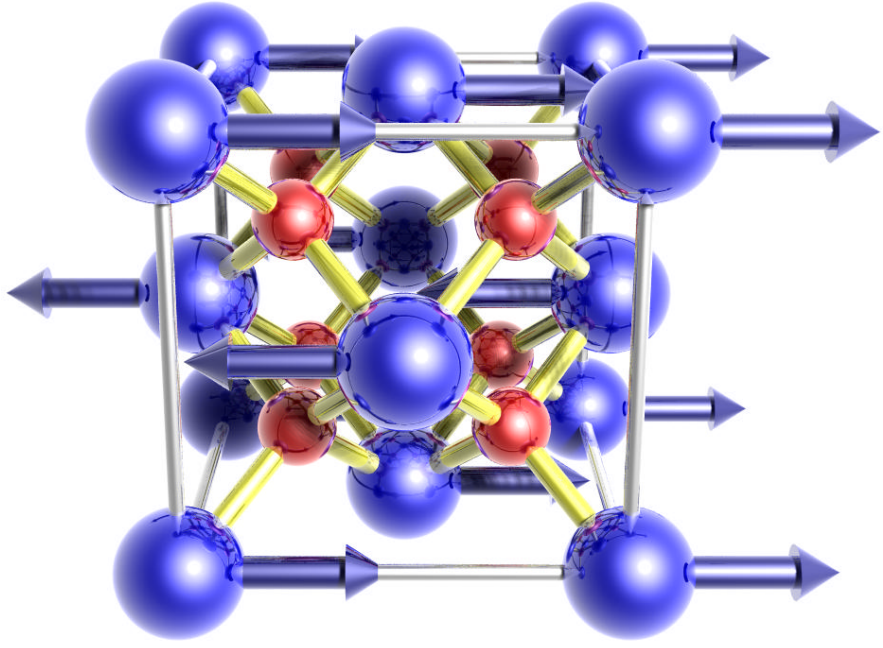
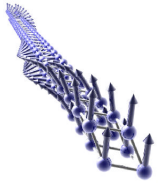
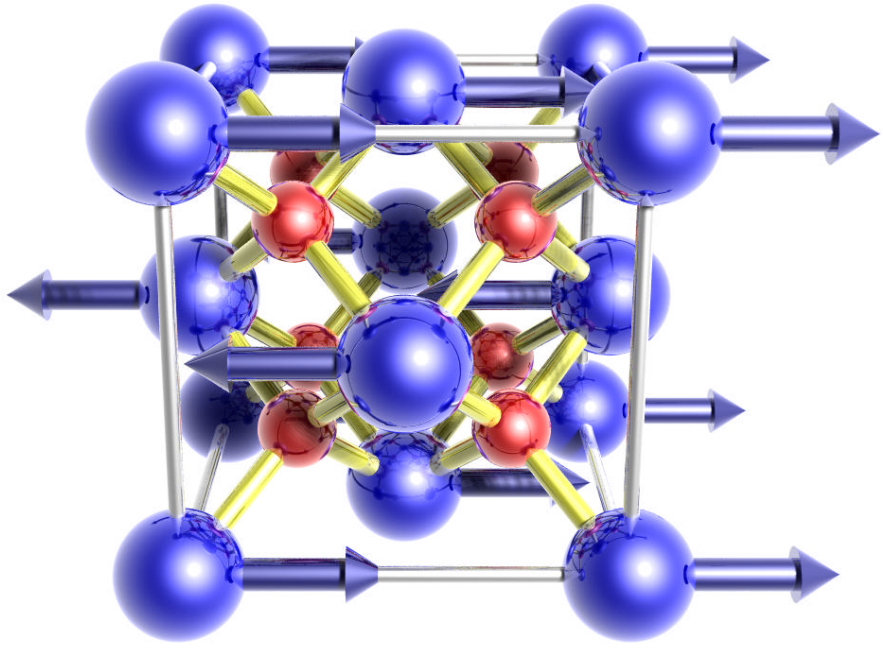
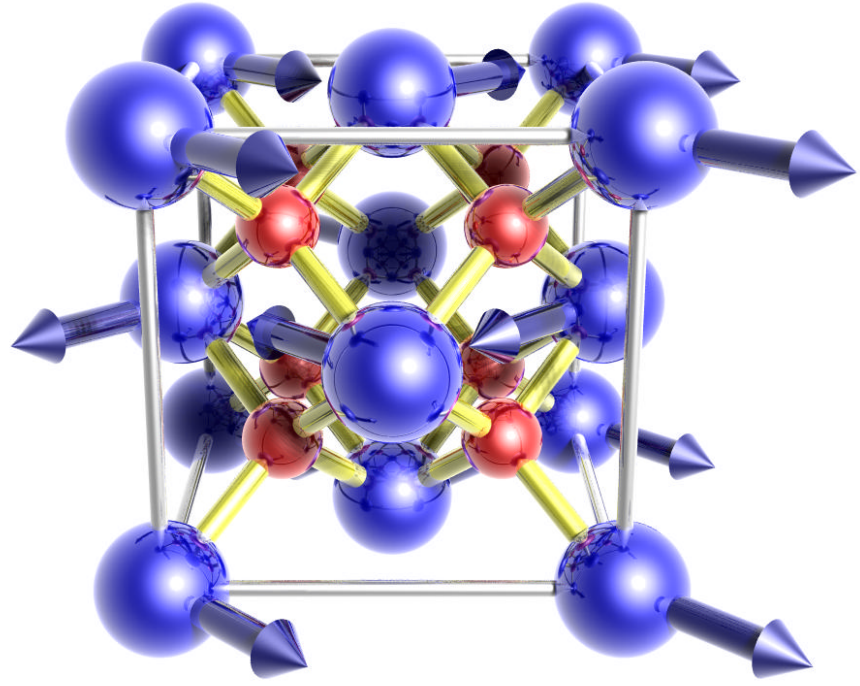
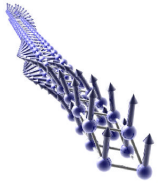






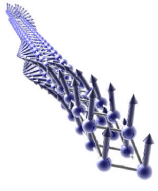






Outline

- theoretical introduction;



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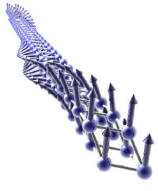
- theoretical introduction;
  - try to place implemented equations in a sequance of approximations begining from Dirac equation



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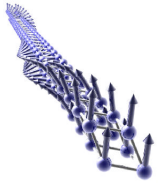
- important details of implementations;



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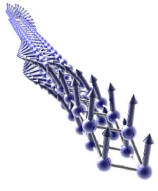
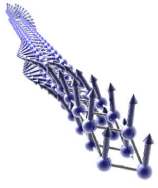


Outline

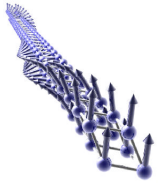
- theoretical introduction;
  - try to place implemented equations in a sequance of approximations begining from Dirac equation
- important details of implementations;
- examples of calculations;
- how to run the code?;



Dirac Hamiltonian

Spin as a dynamical variable is of great importance in magnetism. Thus we start with Dirac Hamiltonian.

$$H_D = c\vec{\alpha} \cdot \vec{p} + \beta mc^2 + V$$



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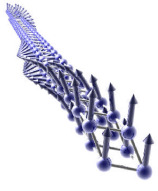
$$H_D = c\,\vec{\alpha}\cdot\vec{p} + \beta mc^2 + V$$

$\vec{\alpha}$ is 3-component vector of 4x4 matrixes:

$$\alpha_k = \begin{pmatrix} 0 & \sigma_k \\ \sigma_k & 0 \end{pmatrix}$$



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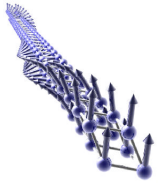
Pauli matrixes:

$$\sigma_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \sigma_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$$

$$\sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$



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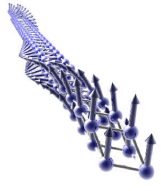

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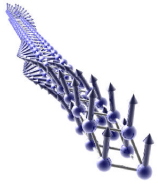
$\mathbf{m}$ - is electron mass, $\mathbf{c}$ - speed of the light

$\mathbf{p}$ - momentum operator

$\mathbf{V}$ - efective potential, which is related to density (DFT)



Dirac equation

$$H_D = c\vec{\alpha} \cdot \vec{p} + \beta mc^2 + V$$

$$H_D \begin{pmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \\ \psi_4 \end{pmatrix} = \varepsilon \begin{pmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \\ \psi_4 \end{pmatrix}$$

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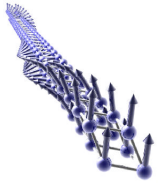

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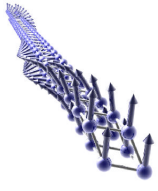
- Hamiltonian is 4x4 matrix operator
- wave function is 4 component vector



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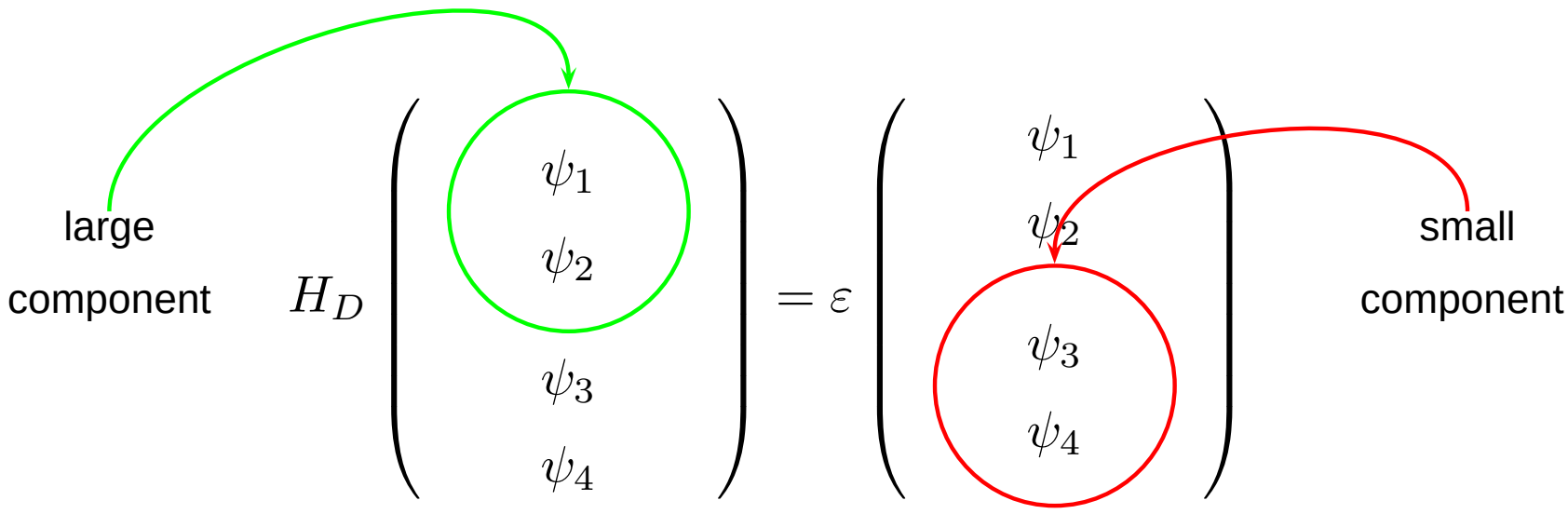

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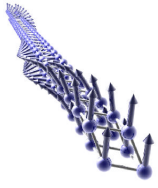

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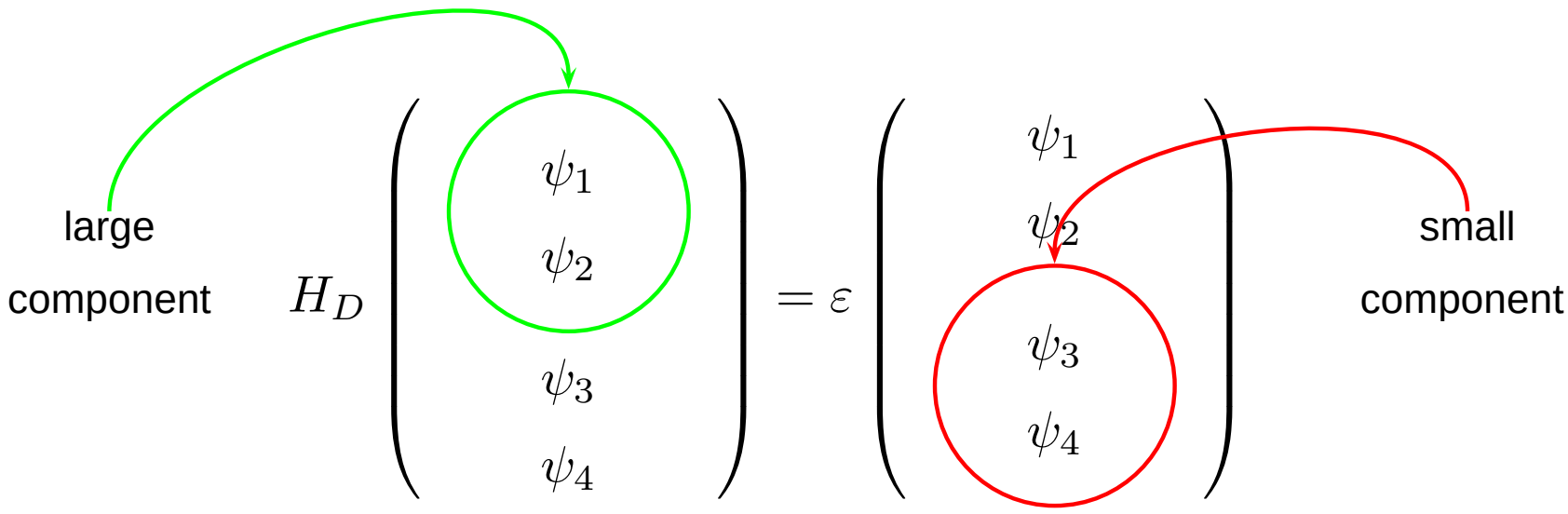
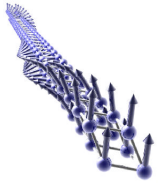
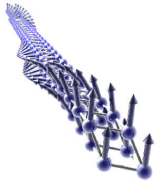


- neglecting contribution of small component to the charge density, one can derive 2x2 Pauli like Hamiltonian



Pauli Hamiltonian

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



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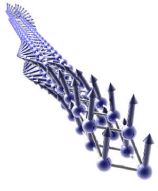

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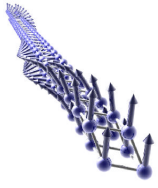

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spin up component

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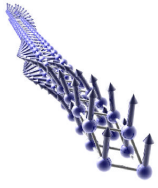
spin down component



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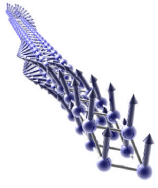
effective electrostatic potential

effective magnetic field

spin-(orbit) interaction term



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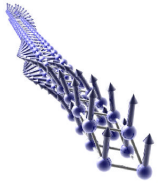

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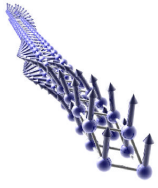
- quite important and difficult to define, describe many body effects



Exchange potential and field

In DFT V_{xc} and $\vec{B}_{xc}$ are defined by:

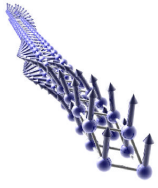
$$V_{xc} = \frac{\partial E_{xc}\left(n, \vec{m}\right)}{\partial n}, \qquad \vec{B}_{xc} = \frac{\partial E_{xc}\left(n, \vec{m}\right)}{\partial \vec{m}}$$

In local density approximation (LDA):

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

what results in:

$$V_{xc} = \epsilon_{xc}\left(n, m\right) + n \frac{\partial \epsilon_{xc}\left(n, m\right)}{\partial n}, \qquad \vec{B}_{xc} = n \frac{\partial \epsilon_{xc}\left(n, m\right)}{\partial m} \hat{m}$$



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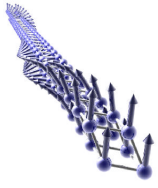
- exchange field is parallel to local magnetisation density vector



Non-collinear case

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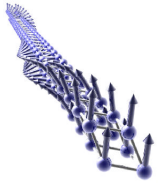
If we take everything, and also allow magnetisation to vary its direction from point to point, we will end up with 2x2 Hamiltonian:

$$\begin{pmatrix} -\frac{\hbar^2}{2m}\nabla^2 + V_{eff} + \mu_B \vec{\sigma} \cdot \vec{B}_z + \ldots & \mu_B \left(B_x - iB_y\right) + \ldots \\ \mu_B \left(B_x + iB_y\right) + \ldots & -\frac{\hbar^2}{2m}\nabla^2 + V_{eff} - \mu_B \vec{\sigma} \cdot \vec{B}_z + \ldots \end{pmatrix} \varphi = \varepsilon \varphi$$



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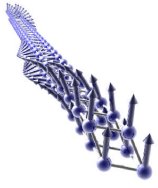
- solutions are non-pure spinors

$$\varphi = \begin{pmatrix} \psi_\uparrow \\ \psi_\downarrow \end{pmatrix}, \quad \psi_\uparrow, \ \psi_\downarrow \neq 0$$

this is non-collinearity



Collinear case

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If we ignore only spin-orbit term, and allow magnetisation to point in one direction (z), resulting Hamiltonian will be diagonal in spin-space.

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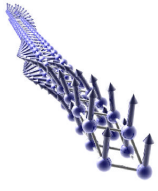

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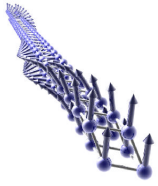
$$\varphi_\uparrow = \begin{pmatrix} \psi_\uparrow \\ 0 \end{pmatrix}, \quad \varphi_\downarrow = \begin{pmatrix} 0 \\ \psi_\downarrow \end{pmatrix}$$

$$\varepsilon_\uparrow \neq \varepsilon_\downarrow$$



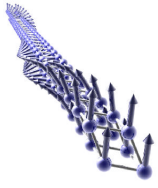
- solutions are pure spinors
- with non-degenerate energies
- collinear magnetism

Non-magnetic case

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

If we ignore spin-orbit term and magnetic exchange field, resulting Hamiltonian will be diagonal in spin-space.

$$\begin{pmatrix} -\frac{\hbar^2}{2m}\nabla^2 + V_{eff} & 0 \\ 0 & -\frac{\hbar^2}{2m}\nabla^2 + V_{eff} \end{pmatrix} \varphi = \varepsilon\varphi$$



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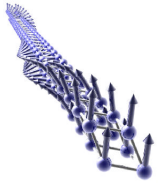
$$\varphi_\uparrow = \begin{pmatrix} \psi \\ 0 \end{pmatrix}, \quad \varphi_\downarrow = \begin{pmatrix} 0 \\ \psi \end{pmatrix}$$

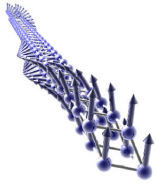
- solutions are pure spinors
- with degenerate energies
- non-magnetic solution

$$\varepsilon_\uparrow = \varepsilon_\downarrow$$



Consequences

Implementation of non-collinearity means that we allow both part of the eigen-spinor ($\psi_\uparrow,\ \psi_\downarrow$) to be non zero at the same time. As a result:



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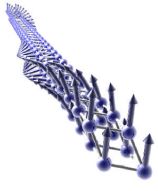

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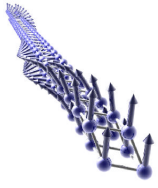

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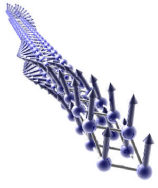
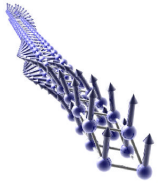
- this means that diagonalisation is 4 times more expensive
- this is usually much more because of losing symmetry operations



Implementations of NCM in LAPW

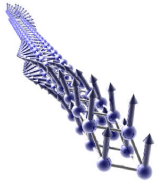
$\psi_\uparrow,\ \psi_\downarrow$ can be non-zero only when basis allows for that.



Implementations of NCM in LAPW

$\psi_\uparrow,\ \psi_\downarrow$ can be non-zero only when basis allows for that.

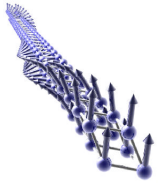
- L. Nordström and D. J. Singh Phys. Rev. Lett. 76, 4420 (1996).
  - pure spinor non polarised basis, given in one global spin coordinate frame,
  - basis must be supplemented by additional local orbitals,
  - magnetisation is a continuous field.



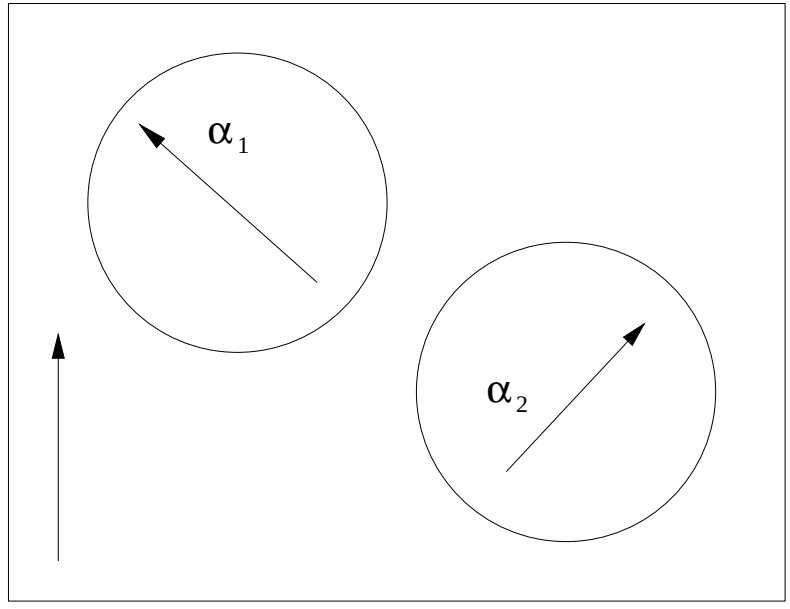
Implementations of NCM in LAPW

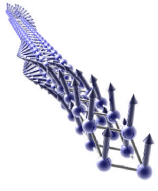
- Ph. Kurz at al. Phys. Rev. B 63, 96401 (2001),
  - interstitial region and each atomic sphere have their own quantisation axis,
  - quantisation axis of a sphere is supposed to point in a direction of average magnetisation,
  - basis functions are pure spinor in interstitial region,
  - inside spheres non-pure spinors but polarised,
  - atomic moment approximation (AMA).

This is implemented in WIEN2k



Spin coordinate sets





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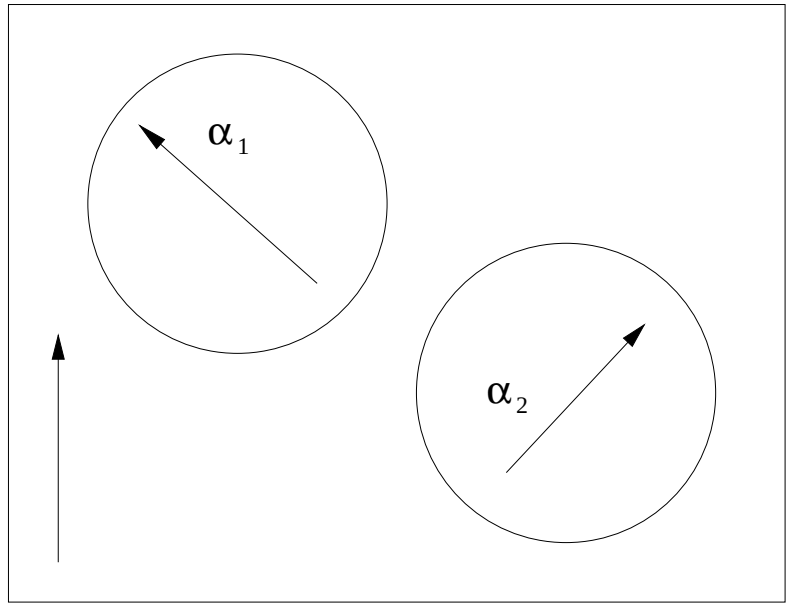
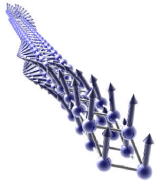
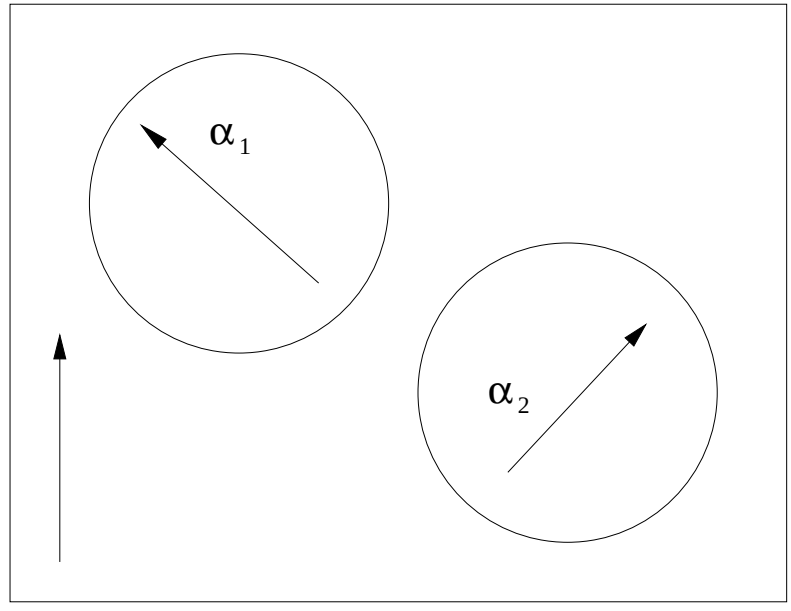


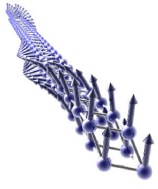
- **interstices:** $\varphi_{\vec{G}\sigma} = e^{i\left(\vec{G}+\vec{k}\right)\cdot\vec{r}}\chi_\sigma$, where $\chi_\sigma = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, or $\begin{pmatrix} 0 \\ 1 \end{pmatrix}$



Basis functions

- **spheres** - combination of pure spinors in a local coordinate frame:
  - the direction of the quantisation axis is along an average magnetisation inside the sphere,





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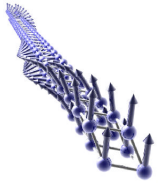
$$\varphi^{APW}_{\vec{G}\sigma}\left(\vec{k}\right) = \sum_{\sigma^\alpha}\sum_{L}\left(A^{\vec{G}\sigma\sigma^\alpha}_{L} u^{\sigma^\alpha}_{l} + B^{\vec{G}\sigma\sigma^\alpha}_{L} \dot{u}^{\sigma^\alpha}_{l}\right) Y_L \chi_{\sigma^\alpha},$$

$$\varphi^{LO}_{\vec{G}\sigma^\alpha} = \left(A^{\vec{G}\sigma^\alpha}_{L} u^{\sigma^\alpha}_{l} + B^{\vec{G}\sigma\sigma^\alpha}_{L} \dot{u}^{\sigma^\alpha}_{l} + C^{\vec{G}\sigma^\alpha}_{L} u^{\sigma^\alpha}_{2,l}\right) Y_L \chi_{\sigma^\alpha}$$

Y_L is a spherical harmonic, L stands for (l, m),
χ_{σ^α} is a spinor given in a local coordinate frame,
u, $\dot{u}$ are radial function and its energy derivative.



Augmentation (...)

$$\varphi^{APW}_{\vec{G}\sigma}\left(\vec{k}\right) = \sum_{\sigma^\alpha}\sum_{L}\left(A_L^{\vec{G}\sigma\sigma^\alpha}u_l^{\sigma^\alpha} + B_L^{\vec{G}\sigma\sigma^\alpha}\dot{u}_l^{\sigma^\alpha}\right)Y_L\chi_{\sigma^\alpha},$$

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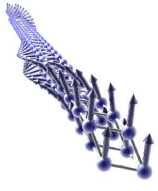
- A_L, B_L, C_L are calculated from sphere boundary conditions.



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- A_L, B_L, C_L are calculated from sphere boundary conditions.
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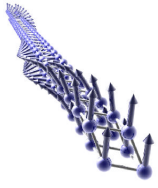

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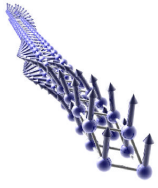

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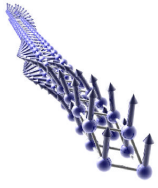
  - for **APW** type of basis $B = 0$ in *PW* and *LO*, and additional local orbital "*lo*" is introduced with $C = 0$, and $B \neq 0$.

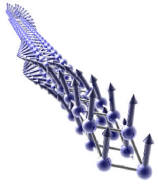


Hamiltonian (...)

- In the **interstitial** region we don't have spin-orbit,
  Hamiltonian is a sum of only kinetic energy and effective potentials:

$$\hat{H} = -\frac{\hbar^2}{2m}\nabla^2 + \hat{V},$$

where $\hat{V}$ combines V_{eff} and B_{eff} in a form of 2x2 potential matrix.



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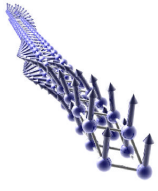
- In the **spheres** w can have everything
  Thus, Hamiltonian is more complicated:

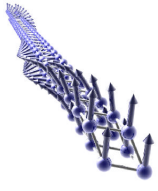
$$\hat{H} = -\frac{\hbar^2}{2m}\nabla^2 + \hat{V} + \hat{H}_{so} + \hat{H}_{orb} + \hat{H}_c$$

  As a result, there are same choices:



Hamiltonian - spheres (...)

- inside sphere potential matrix: $\hat{V} = \hat{V}^d + \hat{V}^{off}$.



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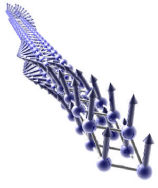

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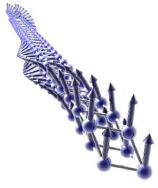
  - **FULL** mode (non-collinearity inside spheres)

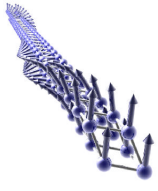
$$\hat{V} = \begin{pmatrix} V_{\uparrow\uparrow} & 0 \\ 0 & V_{\downarrow\downarrow} \end{pmatrix} + \begin{pmatrix} 0 & V_{\uparrow\downarrow} \\ V_{\downarrow\uparrow} & 0 \end{pmatrix}$$



Hamiltonian - spheres (...)

- **SO** (spin-orbit coupling) -

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



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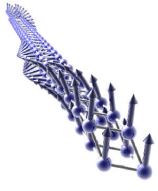
$$\hat{H}_{orb} = \sum_{m,m'} \begin{pmatrix} |m\rangle\, v^{\uparrow}_{mm'}\, \langle m'| & 0 \\ 0 & |m\rangle\, v^{\downarrow}_{mm'}\, \langle m'| \end{pmatrix},$$



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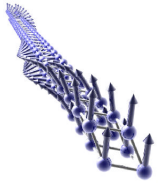
- constrain field ($\vec{B}_c \perp \hat{z}_\alpha$) -

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



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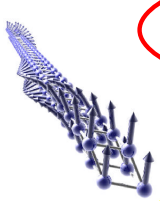
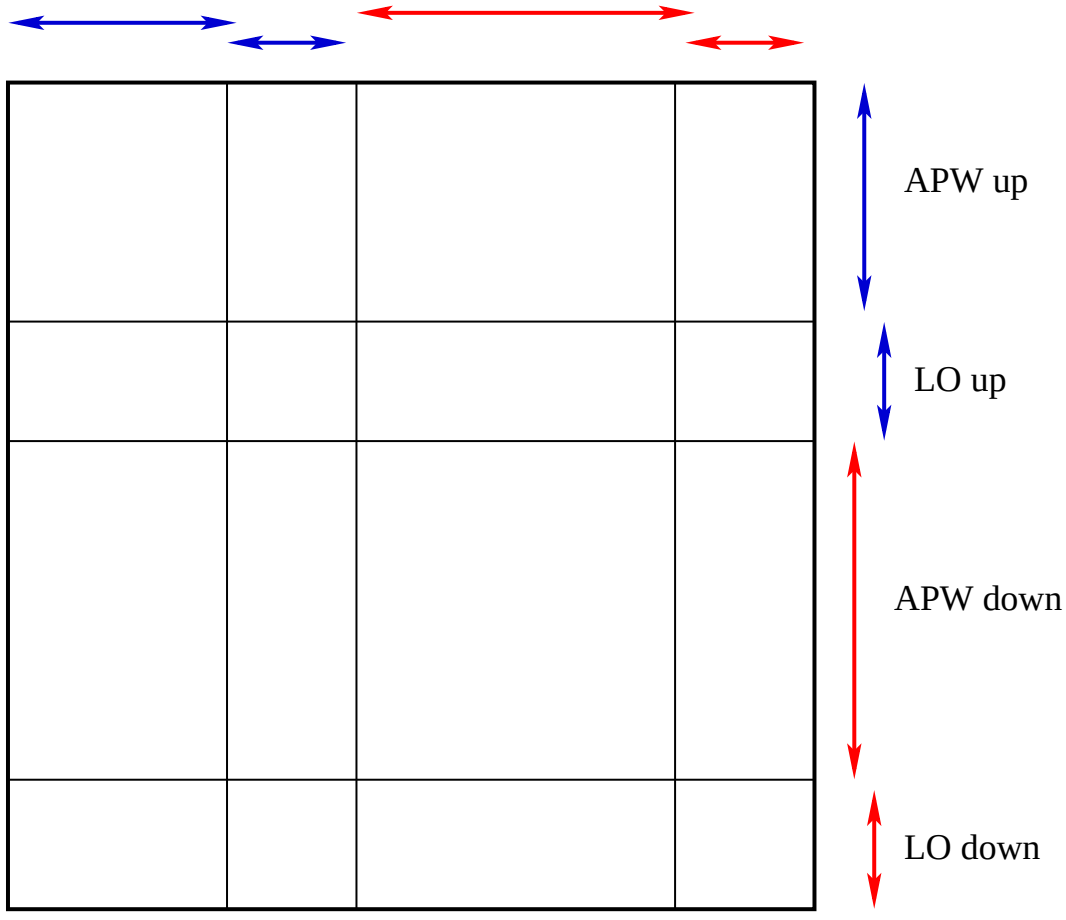


Allocation of the Hamiltonian and overlap matrixes (...)

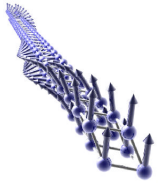


Matrix elements (integrals):

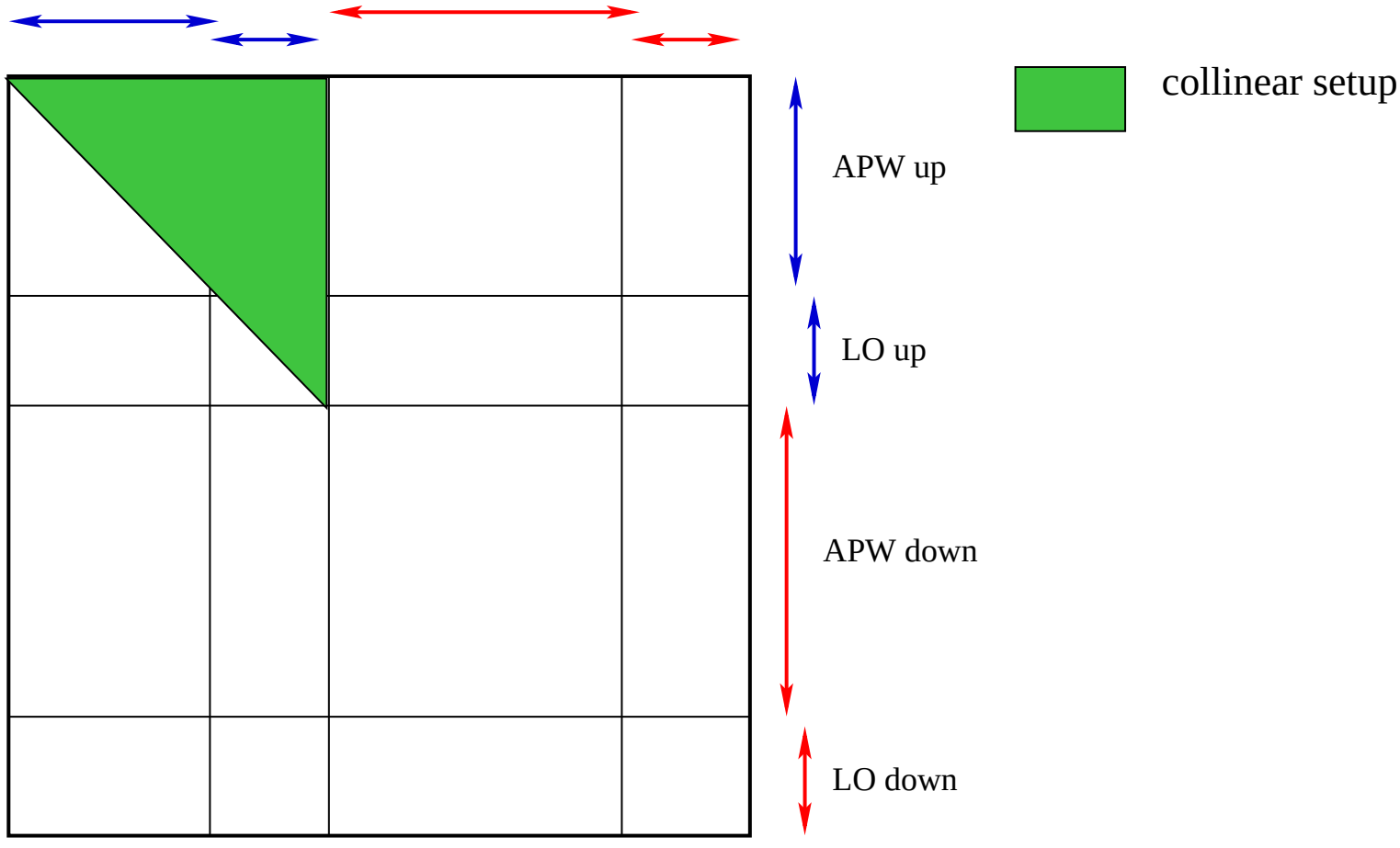
$$H_{\vec{G},\vec{G}'} = \left\langle \psi_{\vec{G}} \middle| \hat{H} \middle| \phi_{\vec{G}'} \right\rangle ,$$

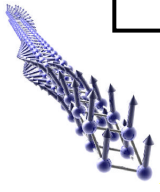
$$S_{\vec{G},\vec{G}'} = \left\langle \psi_{\vec{G}} \mid \phi_{\vec{G}'} \right\rangle ,$$

where ψ and ϕ can be APW or LO, $\uparrow$ or $\downarrow$



Allocation of the Hamiltonian and overlap matrixes (...)





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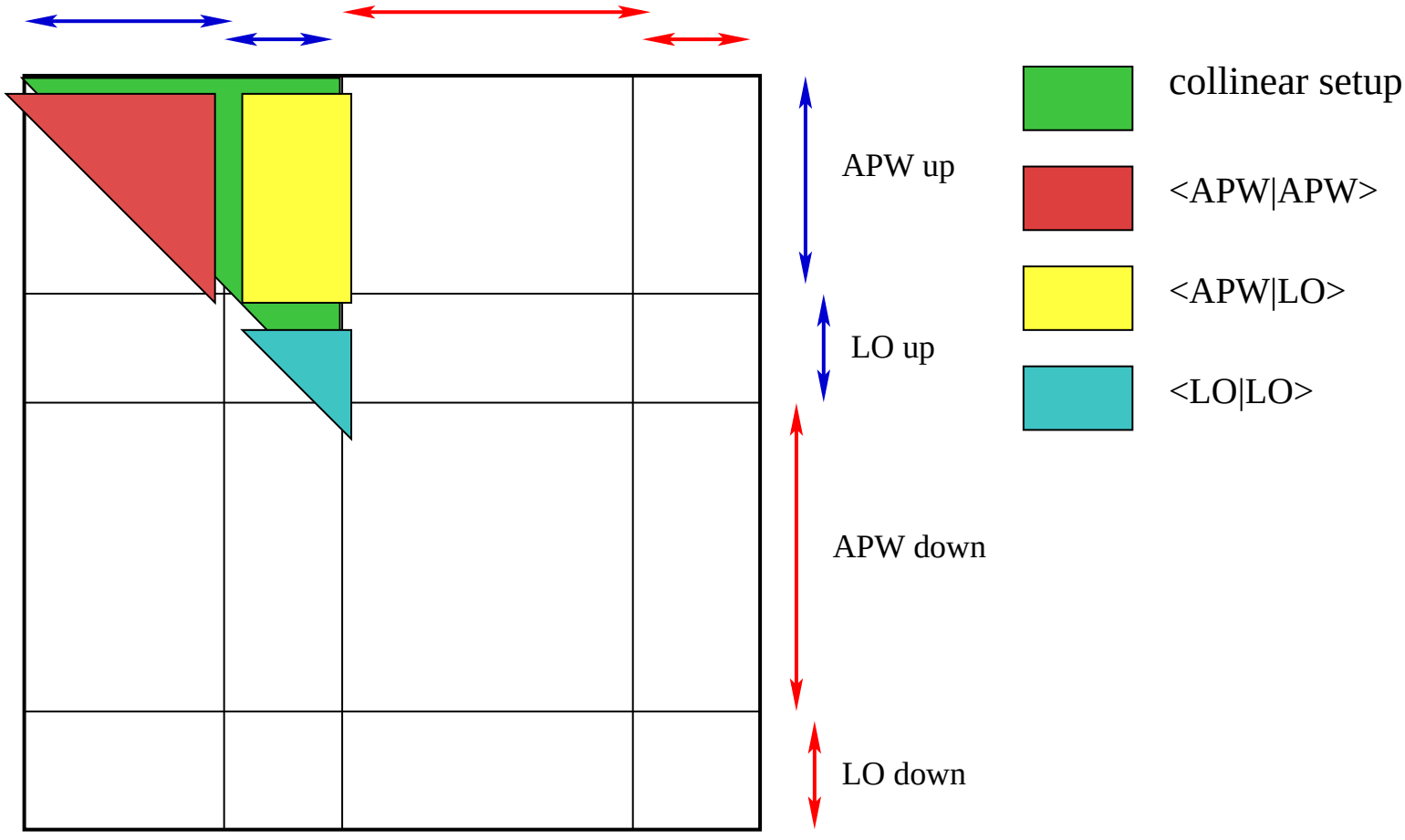

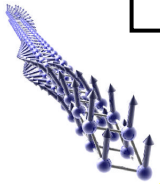

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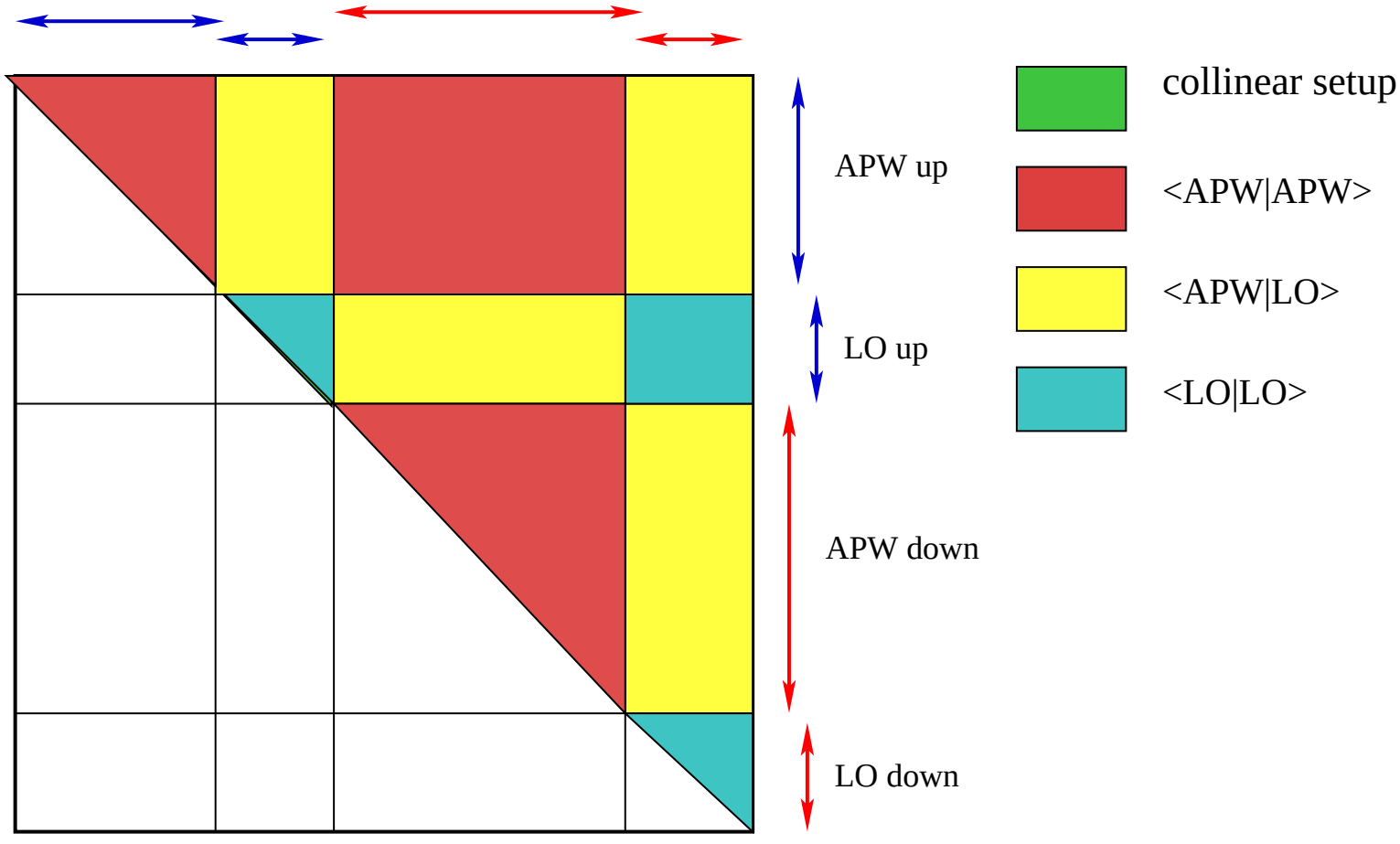
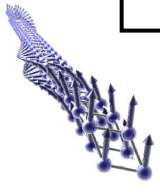
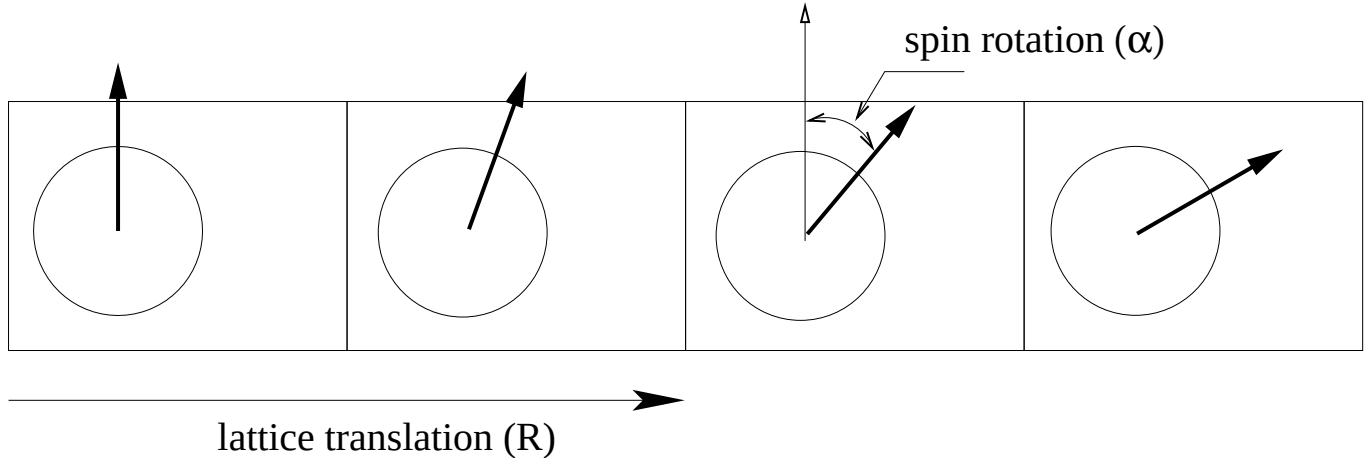




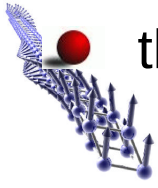
Spin-spirals

- spin-spiral is defined by a vector $\vec{q}$ given in reciprocal space and an angle υ between magnetic moment and rotation axis,



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

$$\vec{m}^n = \vec{m} \left(\cos\left(\vec{q} \cdot \vec{R}^n\right) \sin\upsilon,\ \sin\left(\vec{q} \cdot \vec{R}^n\right) \sin\upsilon,\ \cos\upsilon \right)$$

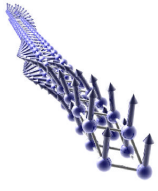


the direction of the rotation axis is arbitrary.

Spin-spirals

- pure translations are obviously not symmetry operations of H,

$$H\left(\vec{r}+\vec{R}^{n}\right) \neq H\left(\vec{r}\right),$$



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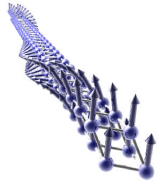
- translations coupled with spin space rotations $T_n = \left\{-\vec{q}\cdot\vec{R}^n \left|\epsilon\right| \vec{R}^n\right\}$ are symmetry operations of H;

$$T_n{}^{\dagger} H\left(\vec{r}\right) T_n = U^{\dagger}\left(-\vec{q}\cdot\vec{R}^n\right) H\left(\vec{r}+\vec{R}^n\right) U\left(-\vec{q}\cdot\vec{R}^n\right),$$



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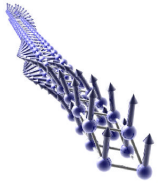
- Group of T_n is Abelian, thus it has one dimensional representations,

$$T_n\psi_k\left(\mathbf{r}\right) = U\left(-\mathbf{q}\cdot\mathbf{R}^n\right)\psi_k\left(\mathbf{r}+\mathbf{R}^n\right) = e^{i\mathbf{k}\cdot\mathbf{R}^n}\psi_k\left(\mathbf{r}\right).$$



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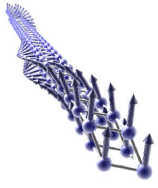


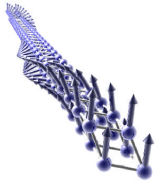
Bloch theorem!!!

Spin-spirals

- Wave function of a spiral structure is of the form:

$$\psi_k(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} \begin{pmatrix} e^{\frac{-i\mathbf{q}\cdot\mathbf{r}}{2}} u_k^{\uparrow}(\mathbf{r}) \\ e^{\frac{i\mathbf{q}\cdot\mathbf{r}}{2}} u_k^{\downarrow}(\mathbf{r}) \end{pmatrix},$$

where $u^{\sigma}(r)$ has translational periodicity.



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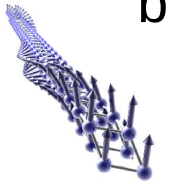
  where $u^{\sigma}(r)$ has translational periodicity.

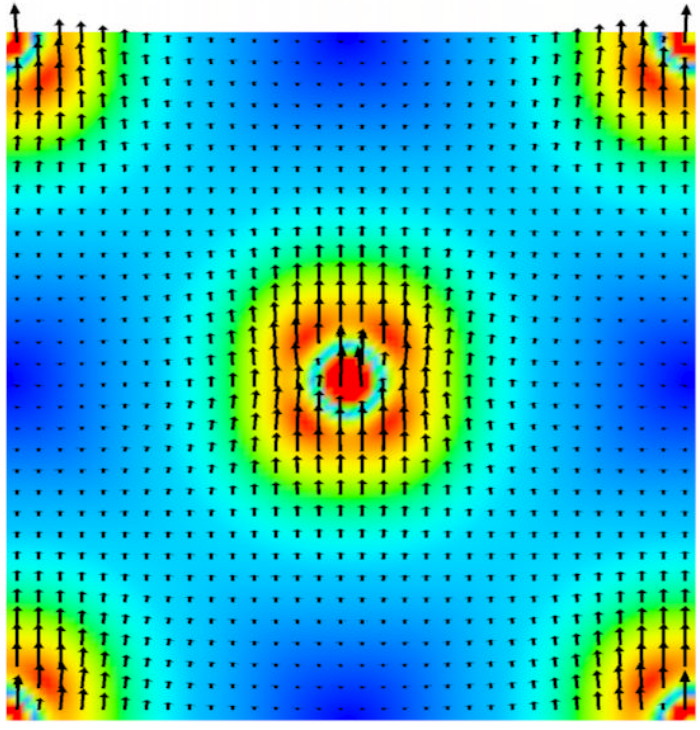
- $\uparrow$ part of a spinor transform with $k - \frac{q}{2}$, $\downarrow$ part of a spinor transform with $k + \frac{q}{2}$. The basis has to be composed using condition:

$$\left|\mathbf{G} + \mathbf{k} \pm \frac{\mathbf{q}}{2}\right| \leq G_{max},$$

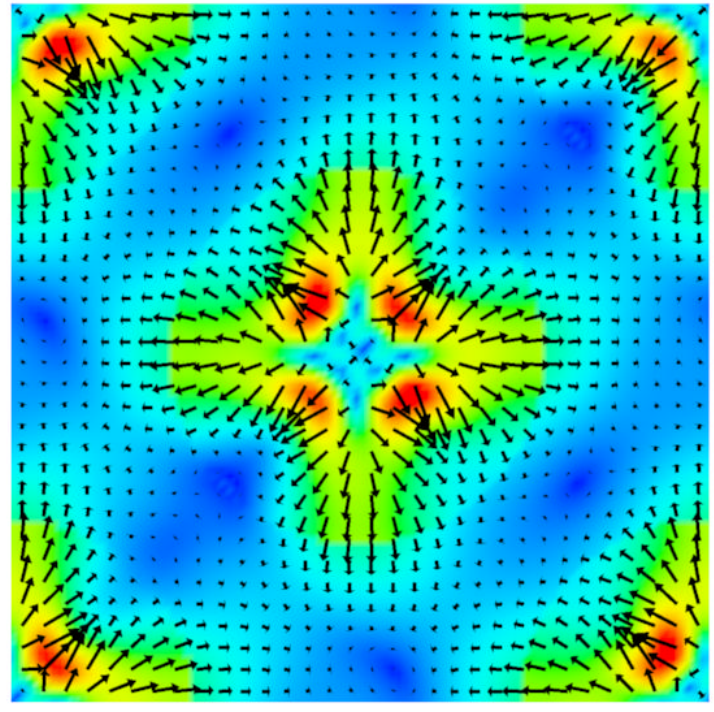
  with "$-$" for $\uparrow$, and "$+$" for $\downarrow$. This can generate different numbers of basis functions for $\uparrow$ and $\downarrow$ spins.



Intra-atomic NCM, fcc Pu

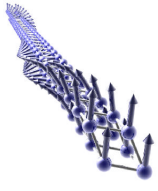


(a) plane $x = 0$

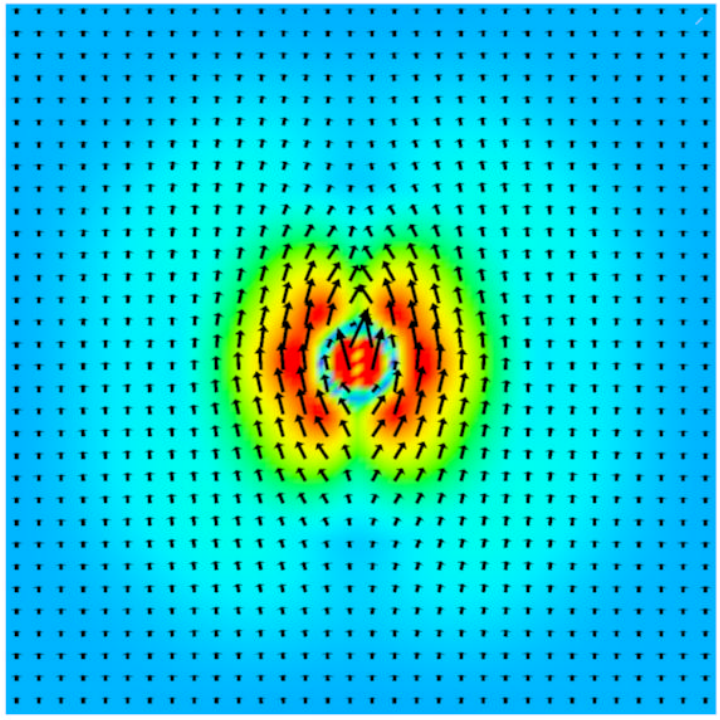


(b) plane $z = 1/10$

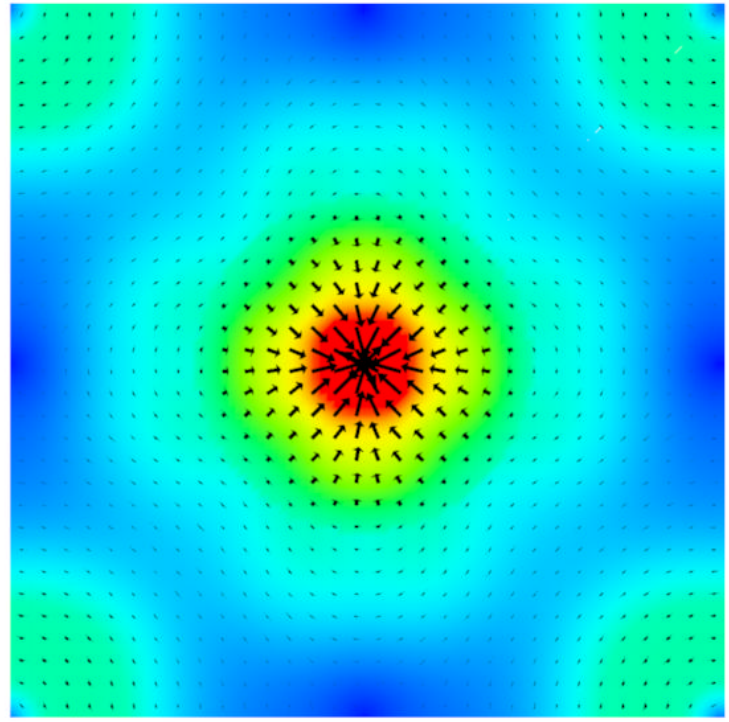
Spin density maps of **fcc Pu**. Calculation in FULL mode with SO. Average momenta point to $\langle 001 \rangle$



Intra-atomic NCM, bcc U

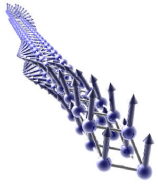


(c) plane $x = 1/2$

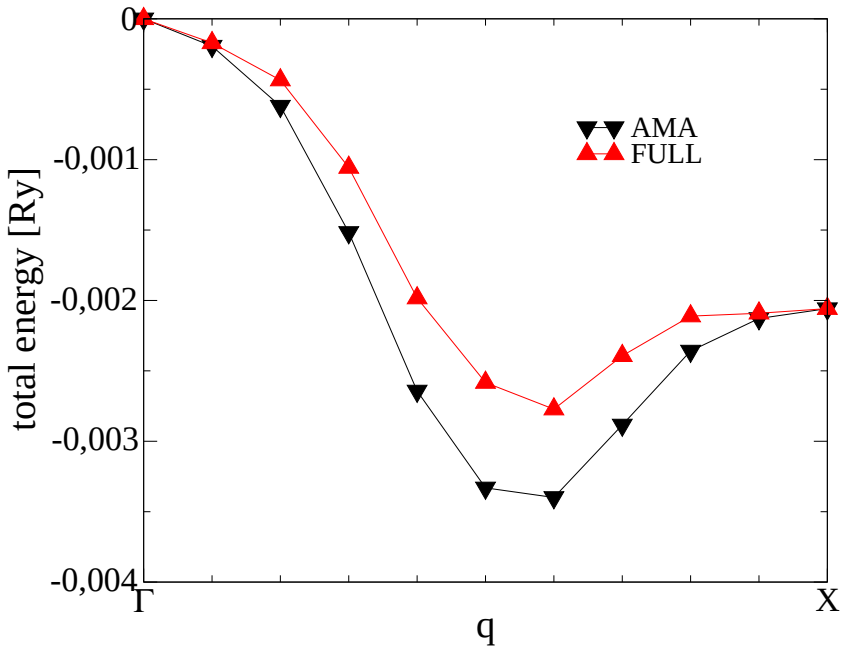


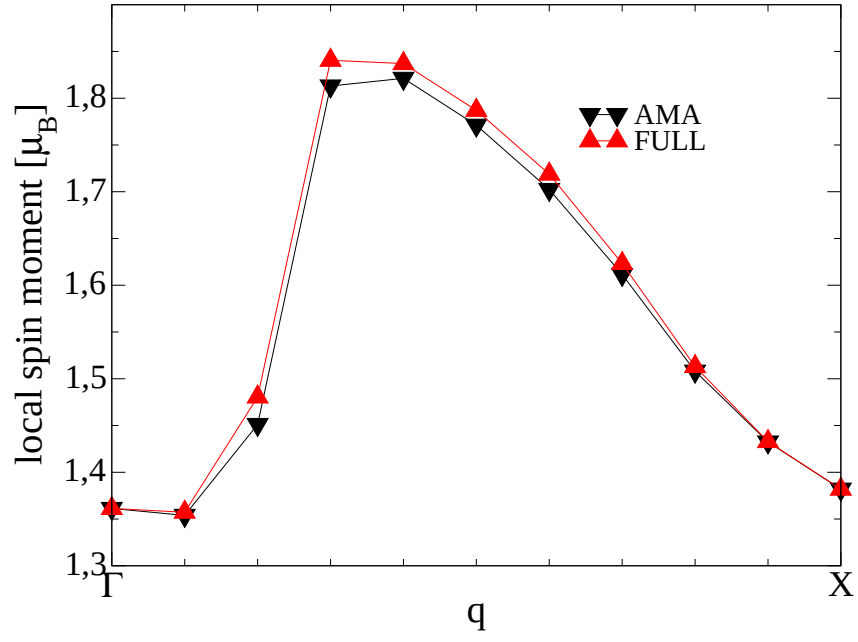
(d) plane $z = 6/10$

Spin density maps of **bcc U** (unit cell size 9 a.u.). Calculation in FULL mode with SO.
Average momenta point to $\langle 001 \rangle$

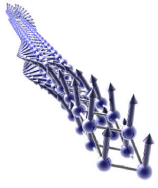


γ Fe, spin spiral





Local spin moment and total energy versus spin-spiral $\vec{q}$ vector.



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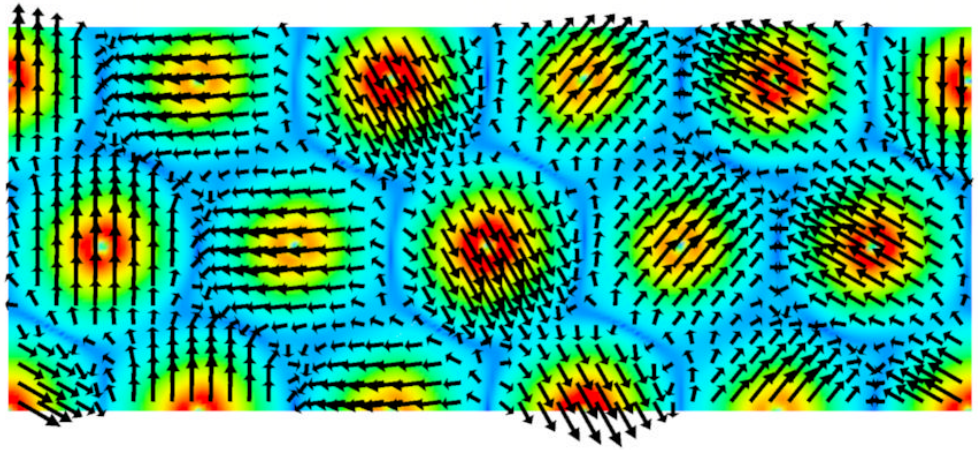
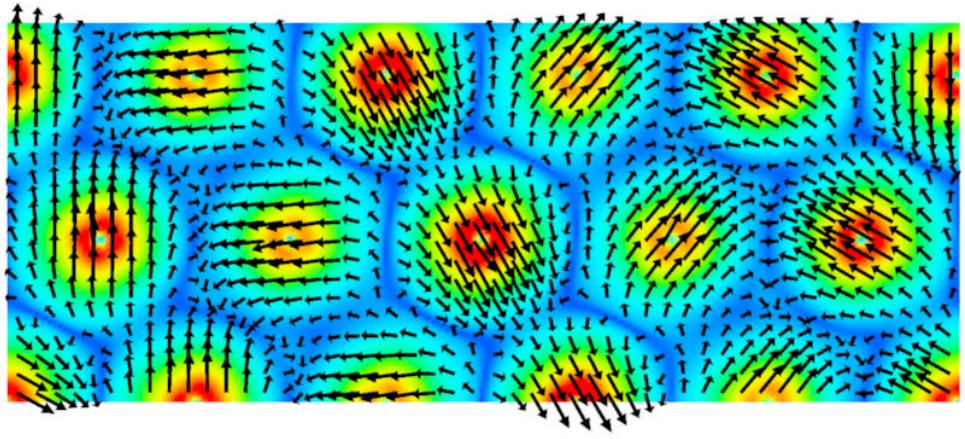
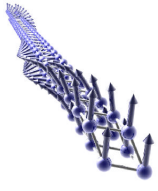


AMA

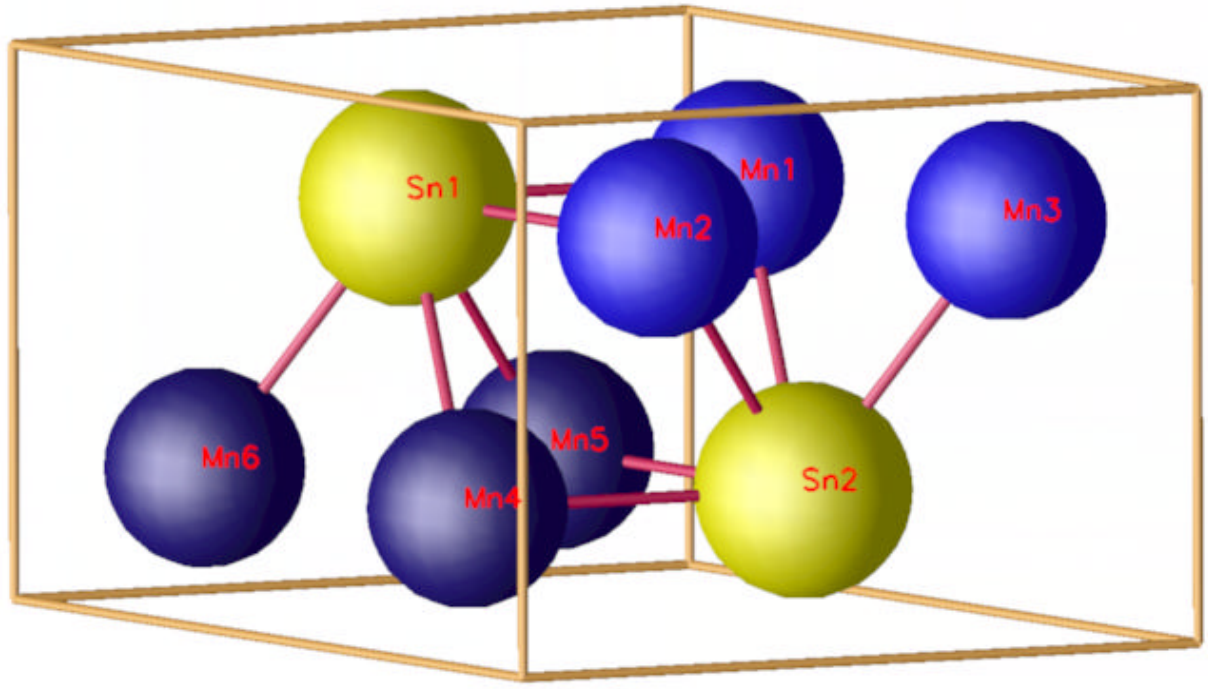


FULL

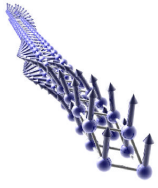


Spin density maps for $q = 0.6$ (0-Γ, 1-X)

Magnetic structure of Mn_3Sn



Chemical structure of Mn_3Sn



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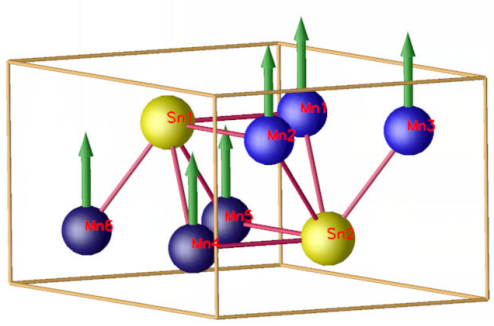
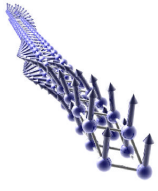


fm



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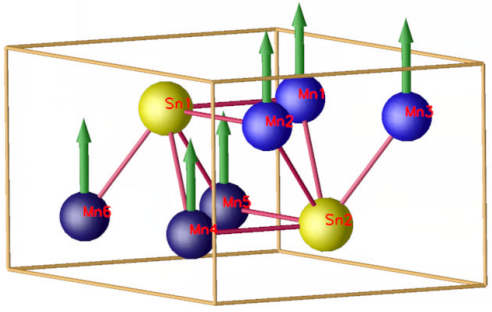

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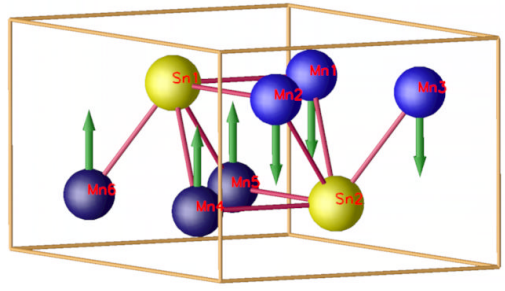
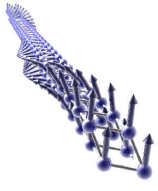


afm



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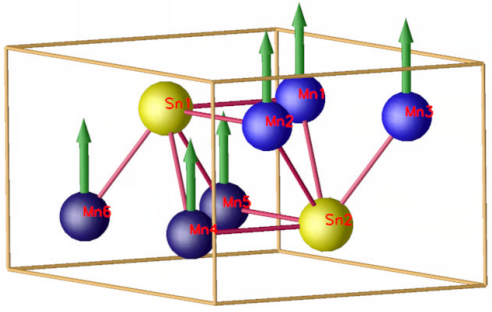

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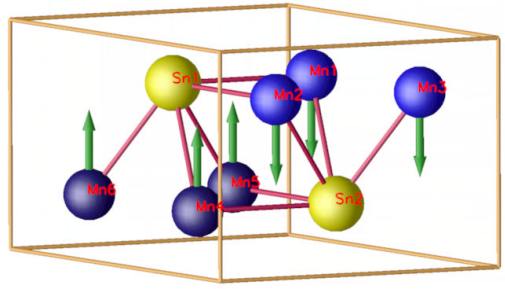

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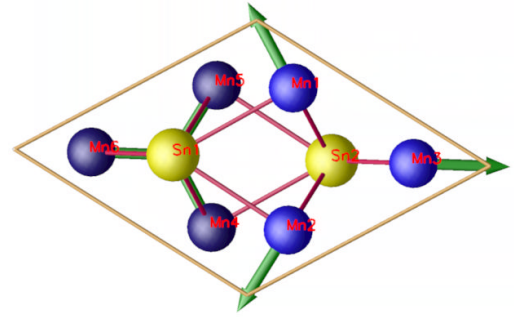
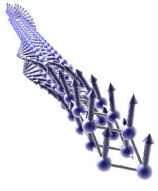


ncm 1



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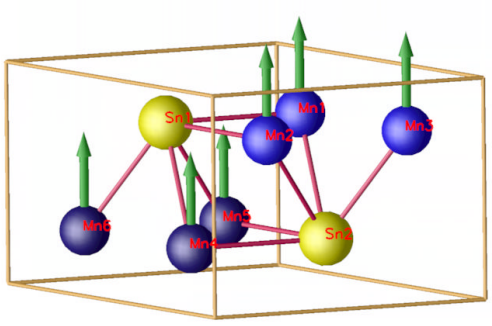
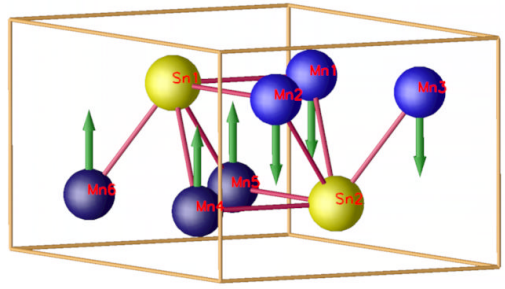


fm



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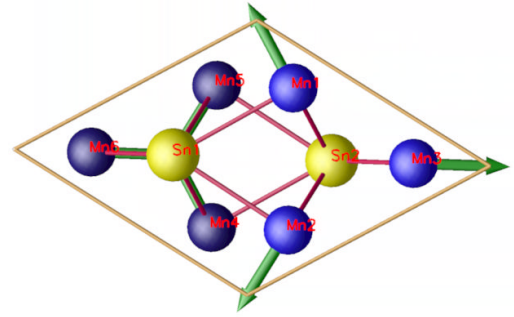

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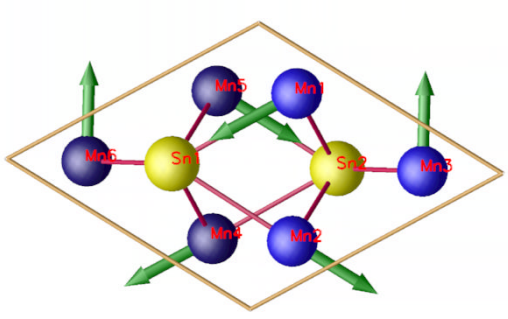
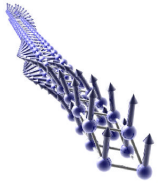


ncm 2



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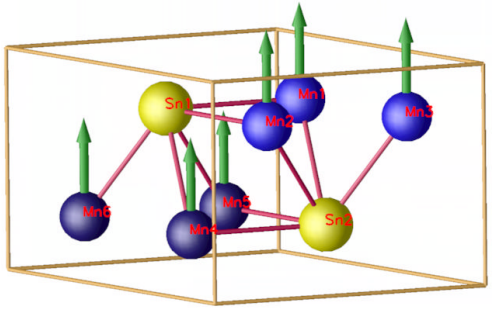

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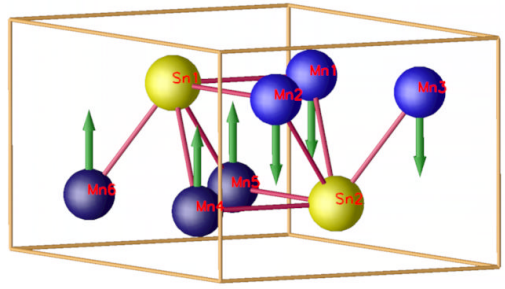

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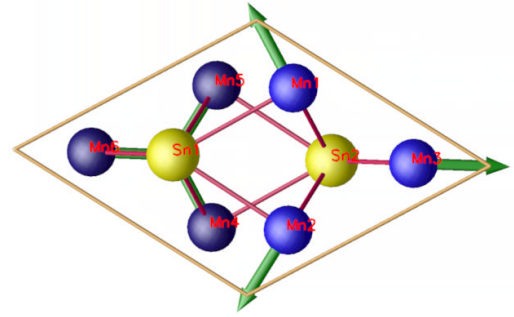
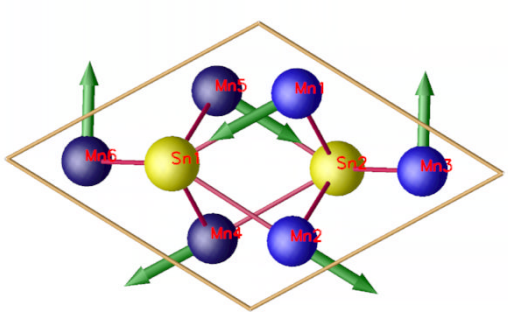
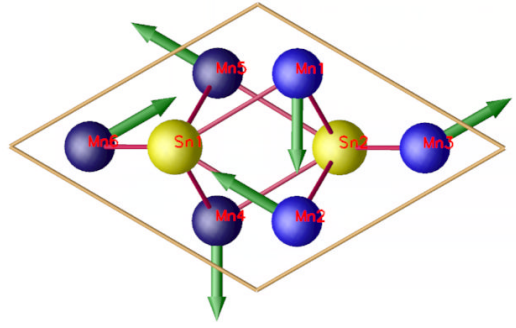


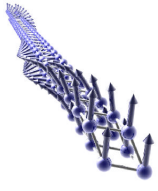
ncm 1



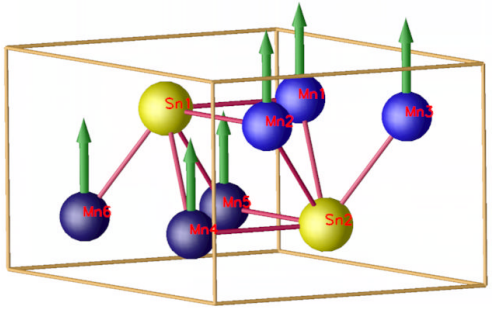
ncm 2



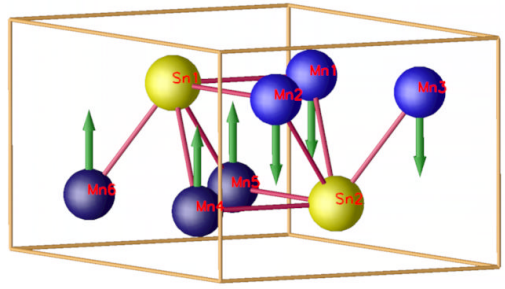
ncm 3



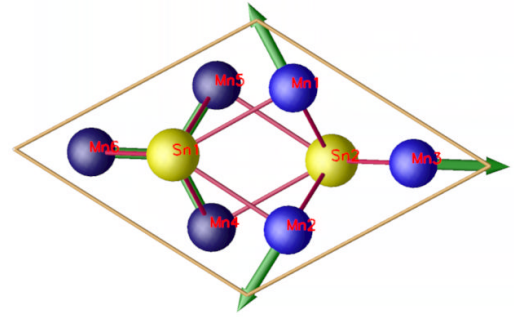
Magnetic structure of Mn_3Sn



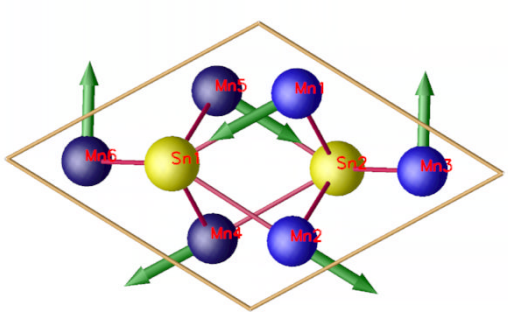
fm



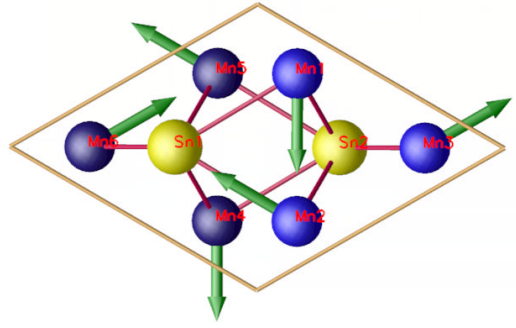
afm



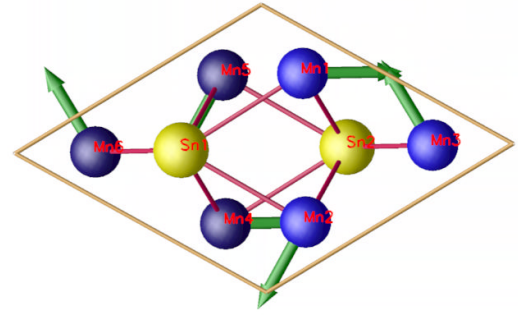
ncm 1



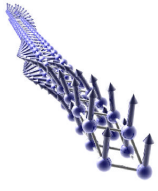
ncm 2



ncm 3



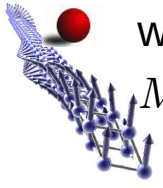
ncm 4



Magnetic structure of Mn_3Sn

	so	fm	afm	ncm 1	ncm 2	ncm 3	ncm 4
$E_{fm} - E\ [Ry]$	-	0.0	0.0131	0.0444	0.0444	0.0444	0.0444
	+	0.0	0.0133	0.0441	0.0439	0.0444	0.0445
$M_s\ [\mu_B]$	-	3.012	2.684	3.037	3.037	3.037	3.037
	+	3.008	2.679	3.034	3.034	3.038	3.037
efg on Mn	-	-1.657	-2.111	-0.894	-0.894	-0.894	-0.894
$[10^{21} V/m^2]$	+	-1.661	-2.119	-0.892	-0.899	-0.891	-0.894
						-0.898	-0.881
hff on Mn	-	-309.9	-153.1	31.2	31.2	31.2	31.2
$[kGauss]$	+	-309.6	-152.9	31.1	31.5	31.5	30.9
						32.2	32.1

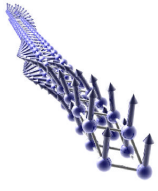
- in *non-so* case all ncm structures are symmetry equivalent
- with *so* ncm structures become inequivalent, additionally for ncm3 and ncm4 Mn_2 and Mn_5 are no longer equivalent to the rest of Mn's



NCM input file

case.inncm:

```
 FULL
 0 0 0
 60.00000 90.00000 0
180.00000 90.00000 0
300.00000 90.00000 0
 60.00000 90.00000 0
180.00000 90.00000 0
300.00000 90.00000 0
  0.00000  0.00000 0
  0.00000 180.00000 0
  0.50000
```

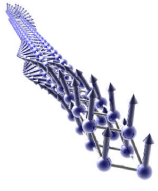


NCM input file

case.inncm:

```
        FULL
        0 0 0
        60.00000 90.00000 0
       180.00000 90.00000 0
       300.00000 90.00000 0
        60.00000 90.00000 0
       180.00000 90.00000 0
       300.00000 90.00000 0
         0.00000  0.00000 0
         0.00000 180.00000 0
         0.50000
```

- first record defines mode of calculation (FULL/AMA)
- second record defines spin-spiral vector;
- next records contain polar angles (ϕ, θ) defining directions of magnetisation in each sphere and optimisation switch;
- last number is a mixing parameter used during calculation of the constrain field;



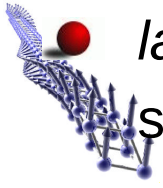
NCM input file

case.inncm:

```
        FULL
        0 0 0
        60.00000 90.00000 0
       180.00000 90.00000 0
       300.00000 90.00000 0
        60.00000 90.00000 0
       180.00000 90.00000 0
       300.00000 90.00000 0
         0.00000  0.00000 0
         0.00000 180.00000 0
         0.50000
```

- first record defines mode of calculation (FULL/AMA)
- second record defines spin-spiral vector;
- next records contain polar angles (ϕ, θ) defining directions of magnetisation in each sphere and optimisation switch;
- last number is a mixing parameter used during calculation of the constrain field;

- *case.inncm* is used in initialisation stage, to lower chemical symmetry due to magnetic moments;
- *lapw1*, *lapw2*, *lapwdm* use it to define quantisation axes for atomic spheres;



How to run it?

- initialisation – looks like in collinear code, but symmetry must be lowered because of magnetic momenta;

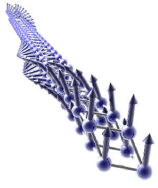
```
x ncmsymmetry
```

- SCF cycle looks like:

```
x lapw0 -ncm [...]
[ x orb -up, x orb -du ]
x lapw1 -ncm [-p -orb -so ...]
x lapw2 -ncm [-p ....]
[ x lapwdm -ncm [-p ...]  ]
x lcore -up
x lcore -dn
x mixer -ncm
```

- SCF job can be run with *runncm* script which is modified version of WIEN2k run script

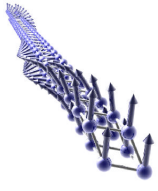
```
runncm [-p -so -orb -cc 0.0001 ...]
```



Acknowledgements

Many thanks to:
Georg Madsen
Peter Blaha
Karlheinz Schwarz

The End



Appendix - augmentation go back

Expansion coefficients $A_L^{\vec{G}\sigma\sigma^\alpha}$ and $B_L^{\vec{G}\sigma\sigma^\alpha}$ are calculated from matching condition:

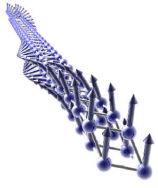
$$e^{i(\vec{G}+\vec{k})\vec{r}}\chi_\sigma^g = \sum_{\sigma^\alpha L}\left(A_L^{\vec{G}\sigma\sigma^\alpha}u_l^{\sigma^\alpha} + B_L^{\vec{G}\sigma\sigma^\alpha}\dot{u}_l^{\sigma^\alpha}\right)Y_L\chi_{\sigma^\alpha}^g\,,$$

where the both spinors are represented in a global coordinate frame. Multiplying both sides by $\left(\chi_{\sigma^\alpha}^g\right)^*$, integrating over spin variable, and comparing to the collinear expression:

$$e^{i(\vec{G}+\vec{k})\cdot\vec{r}} = \sum_{L}\left(A_L^{\vec{G}\sigma^\alpha}u_l^{\sigma^\alpha} + B_L^{\vec{G}\sigma^\alpha}\dot{u}_l^{\sigma^\alpha}\right)Y_L\,.$$

$A_L^{\vec{G}\sigma\sigma^\alpha}$ and $B_L^{\vec{G}\sigma\sigma^\alpha}$ are given by:

$$\begin{aligned} A_L^{\vec{G}\sigma\sigma^\alpha} &= \left(\chi_{\sigma^\alpha}^g\right)^*\chi_\sigma^g A_L^{\vec{G}\sigma^\alpha}\,, \\ B_L^{\vec{G}\sigma\sigma^\alpha} &= \left(\chi_{\sigma^\alpha}^g\right)^*\chi_\sigma^g B_L^{\vec{G}\sigma^\alpha}\,. \end{aligned}$$



Appendix - Hamiltonian setup

go back

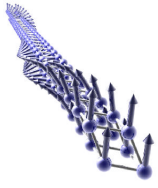
- **interstices** - basis functions are plane waves: $\varphi_{\vec{G}\sigma} = e^{i(\vec{G}+\vec{k})\cdot\vec{r}}\chi_\sigma$, and matrix elements:

$$\left\langle \varphi_{\vec{G}\sigma} \left| \hat{H} \right| \varphi_{\vec{G}'\sigma'} \right\rangle = (V_{\sigma\sigma'}\Theta)_{(\vec{G}-\vec{G}')} + \delta_{\sigma\sigma'}\frac{\hbar}{2m}\left(\vec{G}'+\vec{k}\right)^2\Theta_{(\vec{G}-\vec{G}')}$$

where Θ is a step function, $V_{\sigma\sigma'}$ are components of the potential matrix.

- **spheres** - $\varphi^{PW}_{\vec{G}\sigma} = \sum_{\sigma^\alpha}\phi^{\vec{G}}_{\sigma\sigma^\alpha}\chi_\alpha = \sum_{\sigma^\alpha}\sum_L A_L^{\vec{G}\sigma\sigma^\alpha}u_l^{\sigma^\alpha}Y_L\chi_\alpha$, setup of the Hamiltonian looks like:

$$\begin{aligned}
\left\langle \varphi^{PW}_{\vec{G}\sigma} \mid \hat{H} \mid \varphi^{PW}_{\vec{G}'\sigma'} \right\rangle &= \sum_{\sigma^\alpha}\sum_L A_L^{\sigma^\alpha}\left(\vec{G},\sigma\right)B_L^{\sigma^\alpha}\left(\vec{G}',\sigma'\right), \\
A_L^{\sigma^\alpha}\left(\vec{G},\sigma\right) &= A_L^{\vec{G}\sigma\sigma^\alpha}, \\
B_L^{\sigma^\alpha}\left(\vec{G}',\sigma'\right) &= \sum_{\sigma'^\alpha}\sum_{L'} A_{L'}^{\vec{G}'\sigma'\sigma'^\alpha}\left\langle u_l^{\sigma^\alpha}Y_L \mid H_{\sigma^\alpha\sigma'^\alpha} \mid u_{l'}^{\sigma'^\alpha}Y_{L'}\right\rangle.
\end{aligned}$$



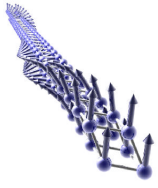
Appendix - density matrix

$$\rho_{\sigma\sigma'} = \frac{1}{2} n I_2 + \sigma \cdot \overrightarrow{m} = \frac{1}{2} \begin{pmatrix} n + m_z & m_x - i\, m_y \\ m_x + i\, m_y & n - m_z \end{pmatrix}$$

- **interstices** ($\rho^{\vec{G}}_{\sigma\sigma'}$)

$$\rho_{\sigma\sigma'}(\vec{r}) = \sum_{\upsilon} \langle \sigma \mid \psi_\upsilon \rangle \langle \psi_\upsilon \mid \sigma' \rangle = \sum_{\vec{G}} \rho^{\vec{G}}_{\sigma\sigma'} e^{i\vec{G}\cdot\vec{r}}$$

  - wave function is generated in real space: $\psi^\sigma_\upsilon(\vec{r}) = \sum_{\vec{G}} c^{\vec{G}}_\upsilon e^{i(\vec{G}+\vec{r})\cdot\vec{r}} \chi_\sigma$, υ indexes band and k-point.
  - $\rho_{\sigma\sigma'}(\vec{r}) = \sum_\upsilon \left(\psi^\sigma_\upsilon(\vec{r})\right)^* \psi^{\sigma'}_\upsilon(\vec{r})$,
  - $\rho_{\sigma\sigma'}(\vec{r})$ is Fourier transformed into $\rho^{\vec{G}}_{\sigma\sigma'}$.



Appendix - density matrix

- **spheres** ($\rho^{L}_{\sigma^{\alpha}\sigma'^{\alpha}}$)

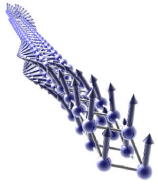
$$\rho_{\sigma^{\alpha}\sigma'^{\alpha}}\left(\vec{r}\right)=\sum_{\upsilon}\left\langle\sigma^{\alpha}\mid\psi_{\upsilon}\right\rangle\left\langle\psi_{\upsilon}\mid\sigma'^{\alpha}\right\rangle=\sum_{L}\rho^{L}_{\sigma^{\alpha}\sigma'^{\alpha}}\left(r\right)Y^{s}_{L}\left(\hat{r}\right)$$

  - $\rho^{L}_{\sigma^{\alpha}\sigma'^{\alpha}}$ are calculated with the following expression:

$$\rho^{L}_{\sigma^{\alpha}\sigma'^{\alpha}}=\sum_{ij}\sum_{l'l''}\left(\sum_{\upsilon}\sum_{m'm''}D^{i\sigma^{\alpha}}_{\upsilon L'}D^{j\sigma'^{\alpha}}_{\upsilon L''}G\left(L,L',L''\right)\right)u^{\sigma^{\alpha}}_{l'}u^{\sigma'^{\alpha}}_{l''},$$

where $D^{i\sigma^{\alpha}}_{\upsilon L}$ (i stands for A, B, C) are given by:

$$\begin{aligned}PW: \quad D^{A,\sigma^{\alpha}}_{\upsilon L} &= \sum_{\sigma}\sum_{G}c^{\vec{G}}_{\upsilon}A^{\vec{G}\sigma\sigma^{\alpha}}_{L}\\ LO,lo: \quad D^{A,\sigma^{\alpha}}_{\upsilon L} &= \sum_{G}c^{\vec{G}}_{\upsilon}A^{\vec{G}\sigma^{\alpha}}_{L}\end{aligned}$$



Appendix - potential matrix

1. local real space diagonalisation:

$$\begin{matrix} \rho^{L}_{\sigma^{\alpha}\sigma'^{\alpha}} \\ \rho^{\vec{G}}_{\sigma\sigma'} \end{matrix} \rightarrow \begin{matrix} \rho_{\sigma^{\alpha}\sigma'^{\alpha}}(\vec{r}) \\ \rho_{\sigma\sigma'}(\vec{r}) \end{matrix} \rightarrow \begin{pmatrix} \rho_{\uparrow}(r) & 0 \\ 0 & \rho_{\uparrow}(r) \end{pmatrix} \rightarrow \begin{matrix} \rho^{L}_{\uparrow},\ \rho^{L}_{\downarrow} \\ \rho^{\vec{G}}_{\uparrow},\ \rho^{\vec{G}}_{\downarrow} \end{matrix}$$

2. generation of collinear potentials:

$$\begin{matrix} \rho^{L}_{\uparrow},\ \rho^{L}_{\downarrow} \\ \rho^{\vec{G}}_{\uparrow},\ \rho^{\vec{G}}_{\downarrow} \end{matrix} \rightarrow \begin{matrix} V^{L}_{\uparrow},\ V^{L}_{\downarrow} \\ V^{\vec{G}}_{\uparrow},\ V^{\vec{G}}_{\downarrow} \end{matrix}$$

3. local de-diagonalisation of collinear potentials in real space:

$$\begin{matrix} V^{L}_{\uparrow},\ V^{L}_{\downarrow} \\ V^{\vec{G}}_{\uparrow},\ V^{\vec{G}}_{\downarrow} \end{matrix} \rightarrow \begin{pmatrix} V_{\uparrow}(r) & 0 \\ 0 & V_{\uparrow}(r) \end{pmatrix} \rightarrow \begin{matrix} V_{\sigma^{\alpha}\sigma'^{\alpha}}(\vec{r}) \\ V_{\sigma\sigma'}(\vec{r}) \end{matrix} \rightarrow \begin{matrix} V^{L}_{\sigma^{\alpha}\sigma'^{\alpha}} \\ V^{\vec{G}}_{\sigma\sigma'} \end{matrix}$$