The all-electron full-potential linearized augmented plane-wave method

Apr 12, 2021 | **Gregor Michalicek** | Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany

MAX DRIVING THE EXASCALE TRANSITION

cecam Centre Européen de Calcul Atomique et Moléculaire

JÜLICH Forschungszentrum

Outline

- **Theoretical background**
  - The FLAPW method and the LAPW basis
  - Separation of core electrons from valence electrons
  - Representation of density and potential
- **Using fleur**
  - The input file generator
  - The inp.xml file
- **Further reading**
- **Conclusion**

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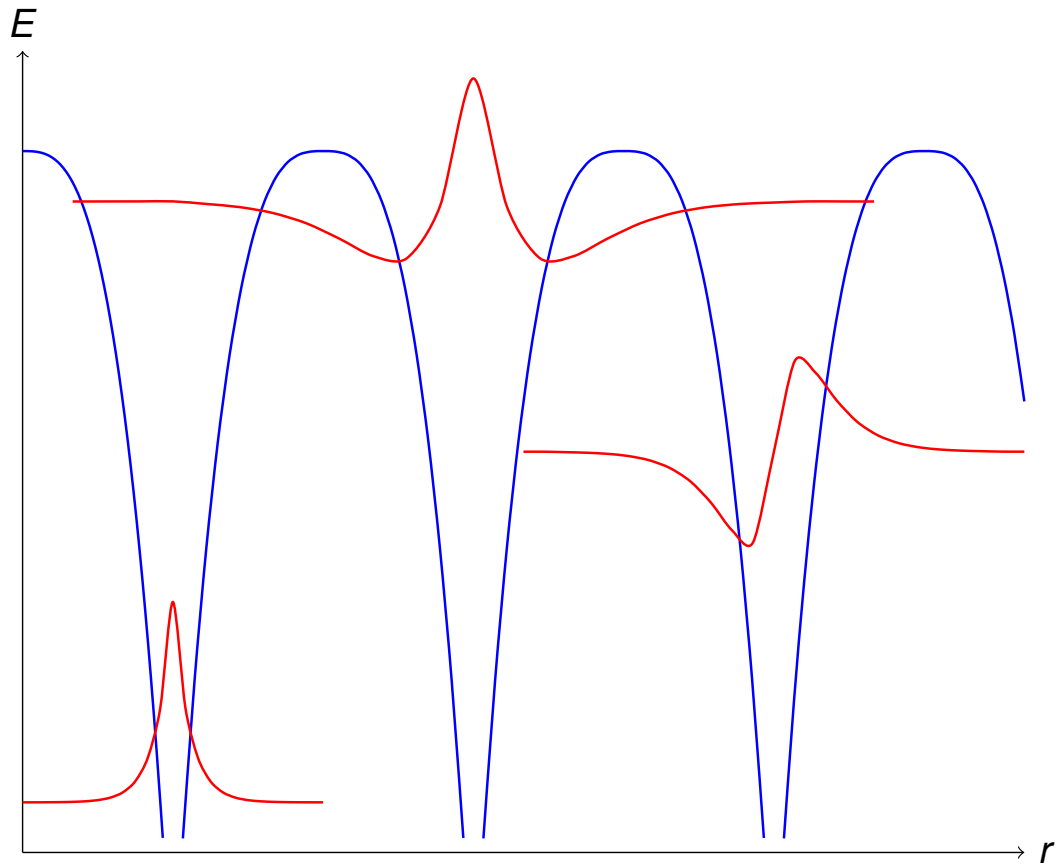
Potential and wave functions in a crystal



Valence electrons

Semicore electrons

Core electrons

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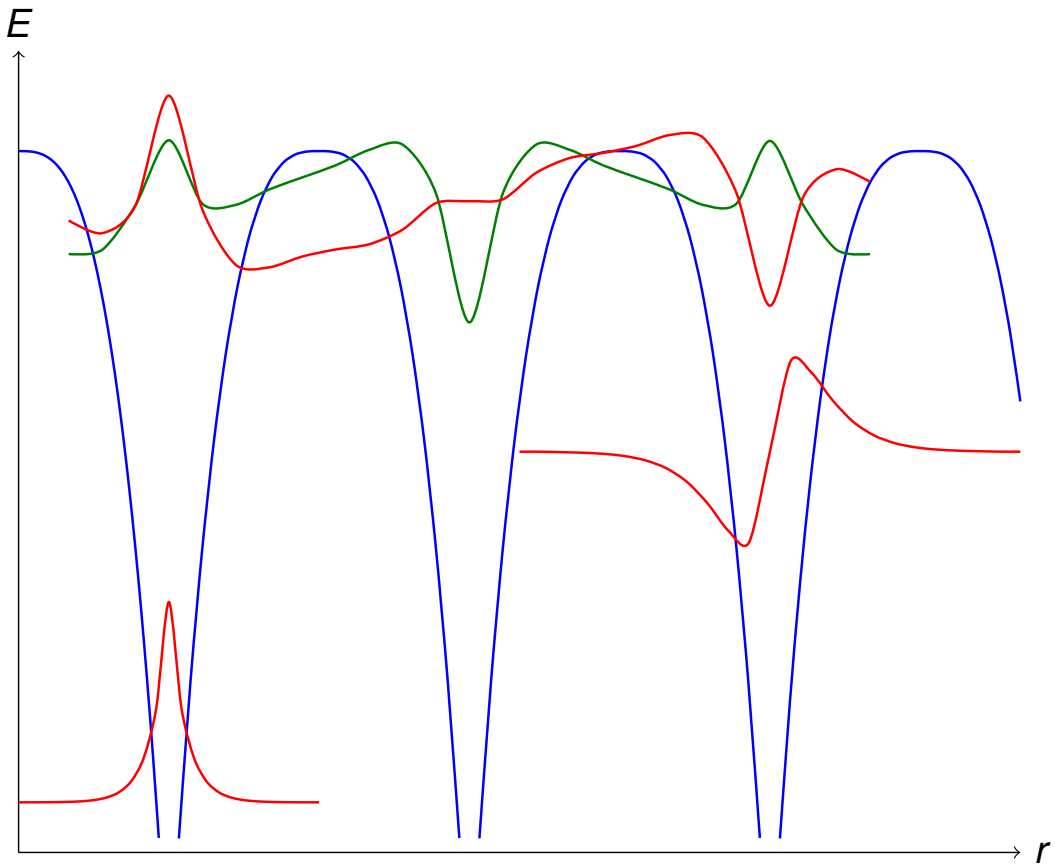

Bloch theorem:

If $V(\mathbf{r}) = V(\mathbf{r} + \mathbf{R})$:

$$\Psi(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}} \cdot u_{\mathbf{k}}(\mathbf{r}),$$

$$u_{\mathbf{k}}(\mathbf{r}) = u_{\mathbf{k}}(\mathbf{r} + \mathbf{R})$$

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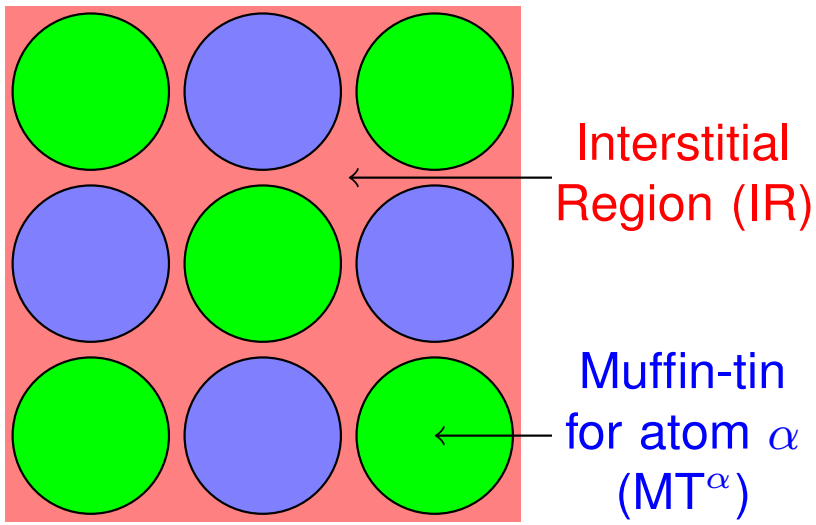
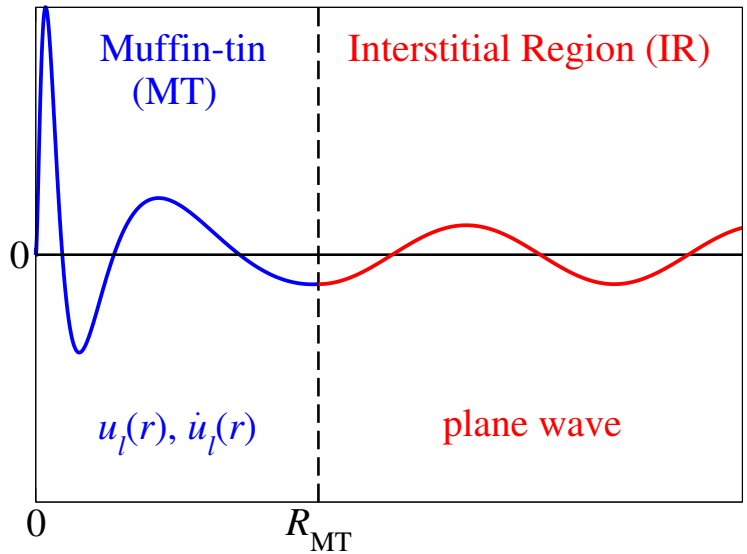
The LAPW basis

- Atom-centered functions in MT spheres matched in value and slope to plane waves in interstitial region (IR)

$$\phi_{\mathbf{kG}}(\mathbf{r}) = \begin{cases} \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{k}+\mathbf{G})\mathbf{r}} & \text{for } \mathbf{r} \in \text{IR} \\ \sum_L \left[a_{\mathbf{kG}}^{L\alpha} u_l^\alpha(r_\alpha, E_l^\alpha) + b_{\mathbf{kG}}^{L\alpha} \dot{u}_l^\alpha(r_\alpha, E_l^\alpha) \right] Y_L(\hat{\mathbf{r}}_\alpha) & \text{for } \mathbf{r} \in \text{MT}^\alpha \end{cases}$$

- u_l^α and $\dot{u}_l^\alpha$ are solutions and energy derivatives for the spherical potential at energy parameters E_l^α





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- u_l^α and $\dot{u}_l^\alpha$ are solutions and energy derivatives for the spherical potential at energy parameters E_l^α

Parameters:

- $K_{\text{max}} = |\mathbf{k} + \mathbf{G}|_{\text{max}}$ reciprocal plane wave cutoff
- l_{max}^α angular momentum cutoff for sphere α
- R_{MT}^α radius for muffin-tin sphere α
- E_l^α energy parameter for u_l^α, $\dot{u}_l^\alpha$

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Orthogonality of LAPW basis functions to core electron states (1)

- $u_l(r)$, $u_l^c(r)$ given by radial Schrödinger equation:

$$\left[-\frac{1}{2}\frac{\partial^2}{\partial r^2} + \frac{l(l+1)}{2r^2} + V_{\text{eff}}^{\text{sphr}}(r)\right] r u_l(r) = E_l r u_l(r) \qquad (1)$$

- Multiply (1) for $u_l(r)$ by $ru_l^c(r)$ and vice versa, subtract the two resulting equations from each other, and integrate:

$$\int_0^{R_{\text{MT}}} -\frac{1}{2} r u_l^c(r) \frac{\partial^2}{\partial r^2} r u_l(r) + \frac{1}{2} r u_l(r) \frac{\partial^2}{\partial r^2} r u_l^c(r) \mathrm{d}r = (E_l - E_l^c) \int_0^{R_{\text{MT}}} u_l(r) r^2 u_l^c(r) \mathrm{d}r$$

- Assumption: $u_l^c(r)\big|_{R_{\text{MT}}} = 0$, $\frac{\partial}{\partial r} u_l^c(r)\big|_{R_{\text{MT}}} = 0$
- We obtain: $0 = \left\langle u_l^c \middle| u_l \right\rangle_{R_{\text{MT}}}$ and analogously $0 = \left\langle u_l^c \middle| \dot{u}_l \right\rangle_{R_{\text{MT}}}$

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Orthogonality of LAPW basis functions to core electron states (2)

- Orthogonality allows to determine core and valence electron energies and wave functions separately from each other
- Core electrons
  - Representation for each atom separately on radial mesh
  - Fully relativistic treatment
- Valence electrons
  - Representation by LAPW basis
  - Scalar-relativistic description in MT spheres
  - Optional inclusion of spin-orbit coupling
- But: assumption $u_l^c(r)\big|_{R_{\mathrm{MT}}} = 0,\ \frac{\partial}{\partial r} u_l^c(r)\big|_{R_{\mathrm{MT}}} = 0$ only approximately fulfilled
  - Semicore states can lead to ghost bands

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The linearization within the LAPW basis

- Description in MT spheres is not systematically improved by increasing the reciprocal cutoff parameter $K_{\max}$

Linearization of solutions u_l at arbitrary energy ϵ

- $u_l^\alpha(r_\alpha, \epsilon) = u_l^\alpha(r_\alpha, E_l^\alpha) + (\epsilon - E_l^\alpha)\dot{u}_l^\alpha(r_\alpha, E_l^\alpha) + \mathcal{O}\left[(\epsilon - E_l^\alpha)^2\right]$
- Due to the restriction to the function space spanned by $u_l^\alpha(r_\alpha, E_l^\alpha)$ and $\dot{u}_l^\alpha(r_\alpha, E_l^\alpha)$ we obtain a linearization error.
- This description is sufficient to obtain accurate results for many materials.

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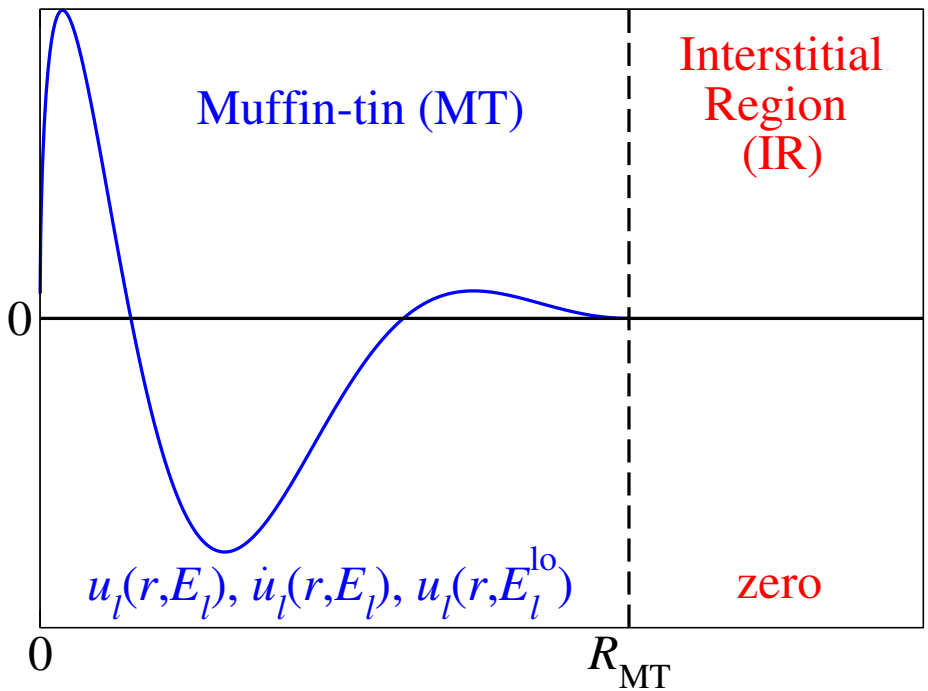
Extending the LAPW basis with local orbitals

Additional basis functions localized in MT spheres

$$\phi_L^{\mathrm{lo}}(\mathbf{r}) = \left[a_L^{\mathrm{lo}} u_l^{\alpha}(r_\alpha, E_l^{\alpha}) + b_L^{\mathrm{lo}} \dot{u}_l^{\alpha}(r_\alpha, E_l^{\alpha}) + c_L^{\mathrm{lo}} u_l^{\alpha}(r_\alpha, E_l^{\mathrm{lo}})\right] Y_L(\hat{\mathbf{r}}_\alpha)$$

- Mainly used to describe semicore states
- Determination of a_L^{lo}, b_L^{lo}, and c_L^{lo} by enforcing zero value and slope at the MT boundary, as well as a normalization condition on the local orbital



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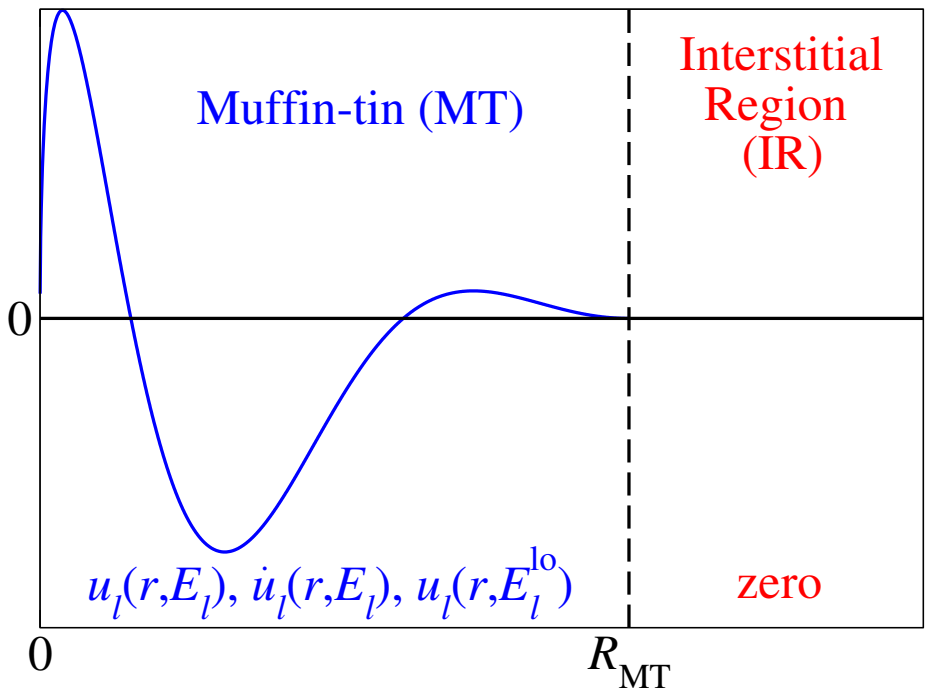
- Semicore states (SCLO)
  - Choose E_l^{lo} to be energy of semicore state
- Unoccupied orbitals (HELO)
  - Choose E_l^{lo} above Fermi energy
- Higher derivative LOs (HDLO)
  - Choose $\ddot{u}_l^{\alpha}(r_\alpha, E_l^{\alpha})$ instead of $u_l^{\alpha}(r_\alpha, E_l^{\mathrm{lo}})$



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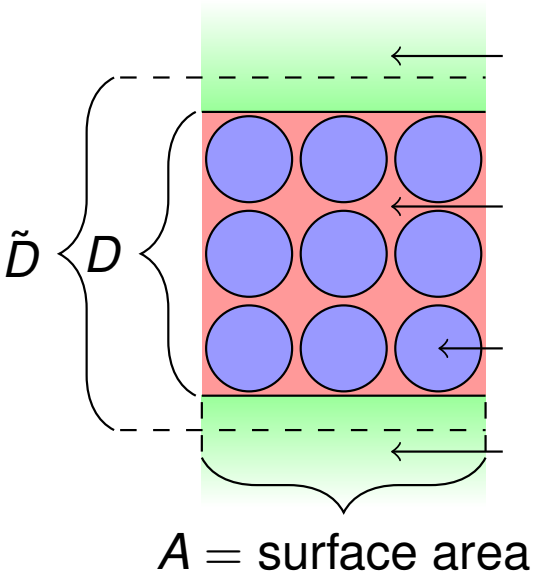
The LAPW basis for films

$$\phi_{\mathbf{k}_\parallel \mathbf{G}}(\mathbf{r}) = \begin{cases} \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{k}_\parallel + \mathbf{G})\mathbf{r}} & \text{for } \mathbf{r} \in \text{IR} \\ \sum_L \left[a^{L\alpha}_{\mathbf{k}_\parallel \mathbf{G}} u^\alpha_l(r_\alpha, E^\alpha_l) + b^{L\alpha}_{\mathbf{k}_\parallel \mathbf{G}} \dot{u}^\alpha_l(r_\alpha, E^\alpha_l) \right] Y_L(\hat{\mathbf{r}}_\alpha) & \text{for } \mathbf{r} \in \text{MT}^\alpha \\ \left[a^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}} u^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}_\parallel}(z, E^{\text{vac}}) + b^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}} \dot{u}^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}_\parallel}(z, E^{\text{vac}}) \right] & \\ \times \frac{1}{\sqrt{A}} e^{i(\mathbf{k}_\parallel + \mathbf{G}_\parallel)\mathbf{r}_\parallel} & \text{for } \mathbf{r} \in \text{VR}^{\text{vac}} \end{cases}$$



Vacuum Region 1 (VR^1)

Interstitial Region (IR)

Muffin-tin (MT)

Vacuum Region 2 (VR^2)

- $u^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}_\parallel}, \dot{u}^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}_\parallel}$: solutions, energy derivatives to vacuum potential at energy parameters E^{vac}
- $G_\perp = 2\pi n / \tilde{D}$

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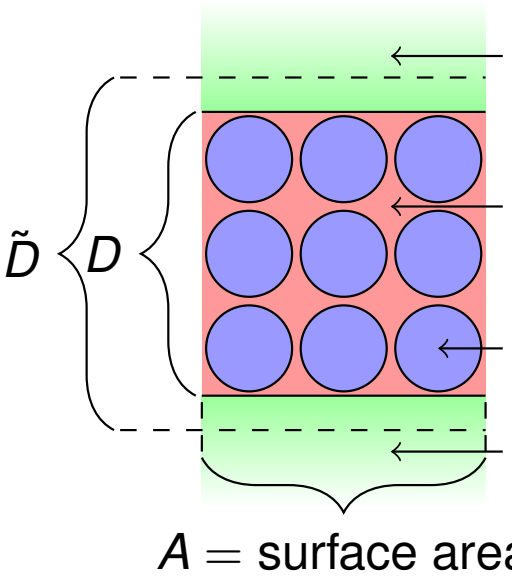
$$\phi_{\mathbf{k}_\parallel \mathbf{G}}(\mathbf{r}) = \begin{cases} \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{k}_\parallel + \mathbf{G})\mathbf{r}} & \text{for } \mathbf{r} \in \text{IR} \\ \sum_L \left[a^{L\alpha}_{\mathbf{k}_\parallel \mathbf{G}} u^\alpha_l(r_\alpha, E^\alpha_l) + b^{L\alpha}_{\mathbf{k}_\parallel \mathbf{G}} \dot{u}^\alpha_l(r_\alpha, E^\alpha_l) \right] Y_L(\hat{\mathbf{r}}_\alpha) & \text{for } \mathbf{r} \in \text{MT}^\alpha \\ \left[a^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}} u^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}_\parallel}(z, E^{\text{vac}}) + b^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}} \dot{u}^{\text{vac}}_{\mathbf{k}_\parallel \mathbf{G}_\parallel}(z, E^{\text{vac}}) \right] & \\ \times \frac{1}{\sqrt{A}} e^{i(\mathbf{k}_\parallel + \mathbf{G}_\parallel)\mathbf{r}_\parallel} & \text{for } \mathbf{r} \in \text{VR}^{\text{vac}} \end{cases}$$



Vacuum Region 1 (VR^1)

Interstitial Region (IR)

Muffin-tin (MT)

Vacuum Region 2 (VR^2)

Parameters:

- D - vacuum boundary
- $\tilde{D}$ - determination of $G_\perp$
- E^{vac} - vacuum energy parameters

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Representation of density and potential

Plane-wave part

$$\rho^{\mathrm{PW}}(\mathbf{r}) = \sum_{\mathbf{G}}^{G_{\max}} \rho_{\mathbf{G}} \cdot e^{i\mathbf{G}\mathbf{r}}$$

- Actually represented by stars
  - Linear combinations of plane waves according to symmetry

MT sphere α

$$\rho^{\alpha}(\mathbf{r}) = \sum_{L}^{L^{\alpha}_{\max}} \rho^{\alpha}_{L}(r_{\alpha})\, Y_L(\hat{\mathbf{r}}_{\alpha})$$

- Actually represented by lattice harmonics
  - Linear combinations of spherical harmonics according to symmetry

Parameters:

- $G_{\max}$ reciprocal plane-wave cutoff for density, potential

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  - The input file generator
  - The inp.xml file
- **Further reading**
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The input file generator

- Fleur uses complex input
  - Input file with default parameters is generated by input file generator **inpgen**
  - Simple structural input needed for inpgen
- Example input for inpgen

```
NaCl bulk

&lattice latsys='fcc' a=10.62026 /

  2
  11 0.0 0.0 0.0
  17 0.5 0.5 0.5
```

- Usage: **inpgen -f myInputFile.txt**
- **-h** write out list of command line options
- Several switches to generation of optional input, e.g., **-noco** for noncollinear magnetism input

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Fleur input: the inp.xml file

```
<?xml version="1.0" encoding="UTF-8" standalone="no"?>
<fleurInput fleurInputVersion="0.34">
   <comment>NaCl bulk</comment>
   <calculationSetup> ... </calculationSetup>
   <cell> ... </cell>
   <atomSpecies> ... </atomSpecies>
   <atomGroups> ... </atomGroups>
   <output> ... </output>
</fleurInput>
```

- Usage: **fleur** or **fleur_MPI**
- **-h** command line option to display all fleur modes

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The inp.xml file - calculationSetup

```
<calculationSetup>
   <cutoffs Kmax="3.6" Gmax="10.8" GmaxXC="10.8"
            numbands="0"/>
   <scfLoop itmax="15" minDistance=".00001"
            imix="Anderson" alpha=".05"/>
   <coreElectrons ctail="T" frcor="F" kcrel="0"/>
   <xcFunctional name="pbe" relativisticCorrections="F"/>
   <magnetism jspins="1"/>
</calculationSetup>
```

+ Optional input, e.g., soc, nocoParams, geometryOptimization

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The inp.xml file - cell

```
<cell>
   <bzIntegration valenceElectrons="16.0" mode="hist"
                  fermiSmearingEnergy=".001">
      <kPointListSelection listName="default"/>
      <xi:include
         xmlns:xi="http://www.w3.org/2001/XInclude"
         href="kpts.xml"> </xi:include>
   </bzIntegration>
   <xi:include xmlns:xi="http://www.w3.org/2001/XInclude"
      href="sym.xml"> </xi:include>
   <bulkLattice scale="1.0">
      <bravaisMatrix>
         <row-1> 0.00000 5.31013 5.31013</row-1>
         <row-2> 5.31013 0.00000 5.31013</row-2>
         <row-3> 5.31013 5.31013 0.00000</row-3>
      </bravaisMatrix>
   </bulkLattice>
</cell>
```


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The inp.xml file - atomSpecies

```
<atomSpecies>
   <species name="Sodium␣(Na)" element="Na"
            atomicNumber="11">
      <mtSphere radius="2.8" gridPoints="925"
                logIncrement=".012"/>
      <atomicCutoffs lmax="10" lnonsphr="8"/>
      <electronConfig flipSpins="F">
         <coreConfig>(1s1/2)</coreConfig>
<valenceConfig>(2s1/2) (2p1/2) (2p3/2) (3s1/2)</valenceConfig
         <stateOccupation state="(3s1/2)" spinUp=".5"
                          spinDown=".5"/>
      </electronConfig>
      <energyParameters s="3" p="3" d="3" f="4"/>
      <lo type="SCLO" l="0" n="2" eDeriv="0"/>
      <lo type="SCLO" l="1" n="2" eDeriv="0"/>
   </species>
   ...
</atomSpecies>
```


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The inp.xml file - atomGroups

```
<atomGroups>
   <atomGroup species="Sodium␣(Na)">
      <relPos label="1">.000000 .000000 .000000</relPos>
      <force calculate="T" relaxXYZ="TTT"/>
   </atomGroup>
   <atomGroup species="Chlorine␣(Cl)">
      <relPos label="2">1.0/2.0 1.0/2.0 1.0/2.0</relPos>
      <force calculate="T" relaxXYZ="TTT"/>
   </atomGroup>
</atomGroups>
```

+ Optional input, e.g., nocoParams

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The inp.xml file - output, relax.xml inclusion

```
<output dos="F" band="F" slice="F">
   <bandDOS minEnergy="-.5*Htr" maxEnergy=".5*Htr"
            sigma=".015" storeEVData="T"/>
</output>
<xi:include xmlns:xi="http://www.w3.org/2001/XInclude"
   href="relax.xml"> <xi:fallback/> </xi:include>
```

+ Optional input, e.g., vacuumDOS, plotting, chargeDensitySlicing

- Full documentation of inp.xml file on www.flapw.de

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Overview on files

with HDF5

- inp.xml, sym.xml, kpts.xml
- enpara (optional)
- cdn.hdf
- mixing_history.*
- input for special calculations
- out, out.xml
- banddos.hdf

without HDF5

- inp.xml, sym.xml, kpts.xml
- enpara (optional)
- cdn1, cdn??, cdnc
- mixing_history.*
- input for special calculations
- out, out.xml

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Further reading

- The FLAPW method
  - Overview book - Singh *et al.*, *Planewaves, Pseudopotentials, and the LAPW Method*, Springer
  - Initial publication - Andersen, PRB **12**, 3060 (1975)
  - FLAPW for films - Krakauer *et al.*, PRB **19**, 1706 (1979)
  - Potential calculation - Weinert, J.Math.Phys. **22**, 2433 (1981)
  - Predecessor (APW) - Slater, Phys.Rev. **51**, 846 (1937)
- Local orbitals
  - SCLOs - Singh, PRB **43**, 6388 (1991)
  - HELOs - Betzinger *et al.*, PRB **83**, 045105 (2011)
  - HDLOs - Friedrich *et al.*, PRB **74**, 045104 (2006)
  - Linearization error - Michalicek *et al.*, CPC **184**, 2670 (2013)

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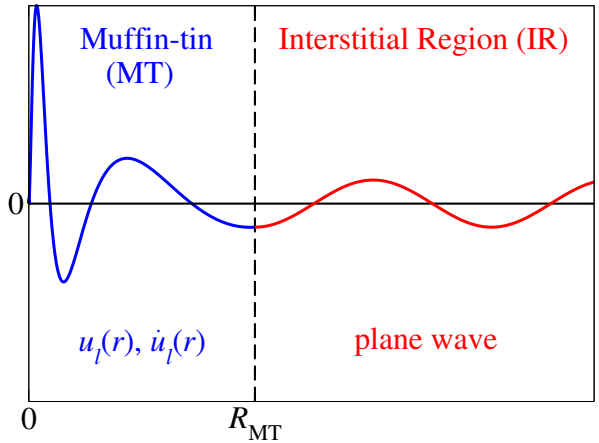
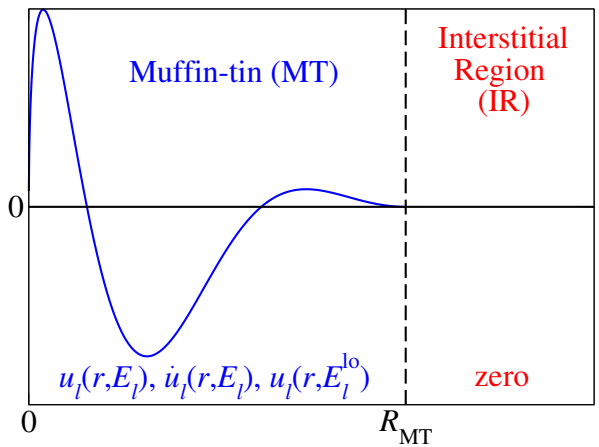
Conclusions

- LAPW basis
- Local orbitals
- The linearization error
- Film setup
- Fleur input and output files
- Not discussed
  - Choosing the parameters
  - General numerical DFT parameters, e.g., k point set, Fermi smearing, ...





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