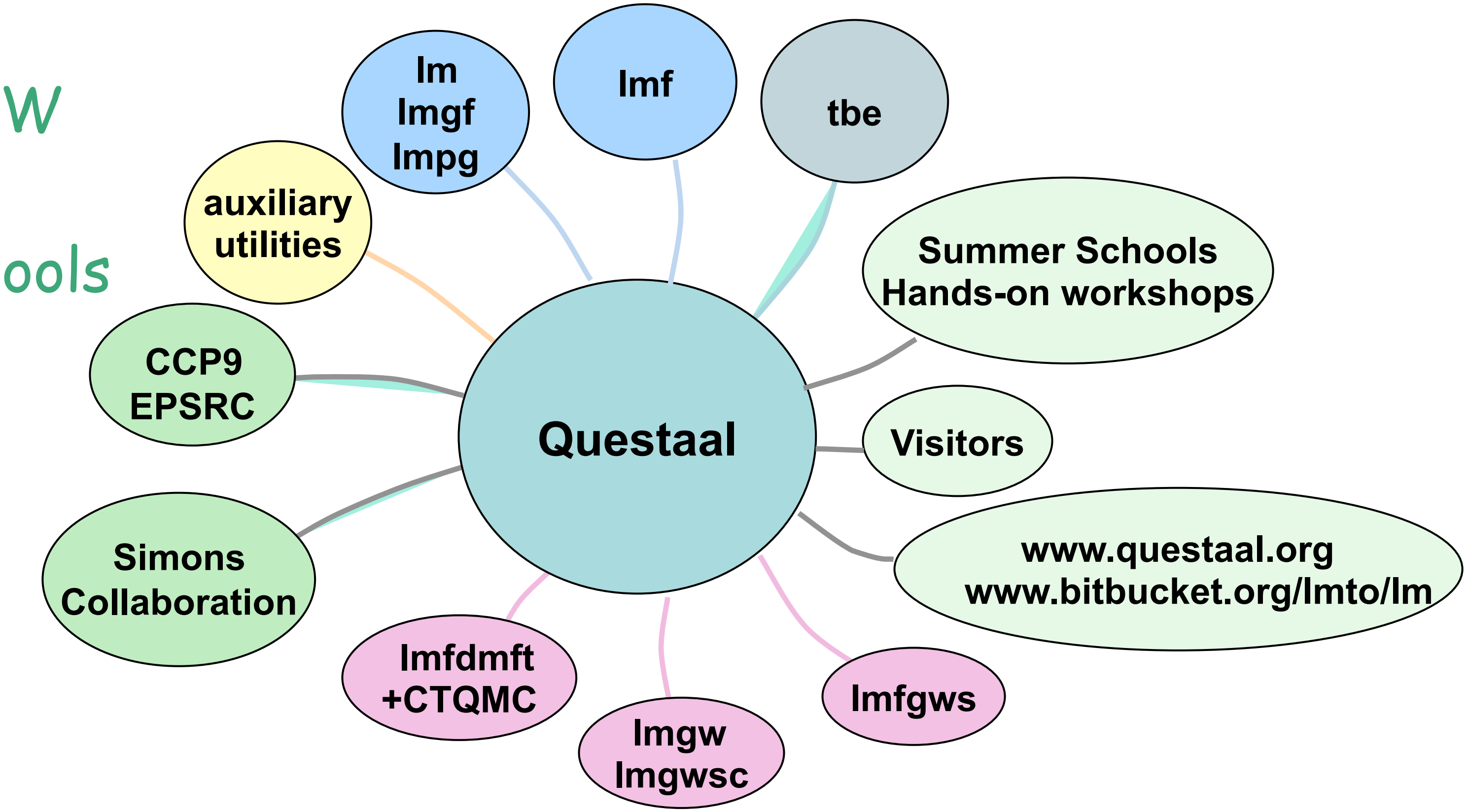
Density Functional Theory and the LMTO method

2017 Questaal Hands-On Course

Mark van Schilfgaarde, King's College London

Questaal is the current CCP9 flagship (www.questaal.org)
It is an integrated electronic structure package that combines:

1. DFT
2. GW, QSGW
3. DMFT
4. Analysis tools



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Outline of Talk

- ✔ How DFT works, general considerations
  Variants of functionals, self-consistency
- ✔ Different implementations of DFT, Survey
- ✔ All-electron methods; how augmentation works
- ✔ KKR Method
- ✔ Linearization ⇒ energy-independent basis
- ✔ The LMTO-ASA approximation
- ✔ Survey of some features of the ASA suite lm, lmgf, lmpg
- ✔ lmf : All-electron Full Potential method

Density-Functional Theory

Hohenberg and Kohn (1964) proved that the total energy E can be determined solely from the density. The total energy is written:

$$E_{HK}[n] = T_s[n] + \underbrace{\int d^3r\, n(\mathbf{r}) V_{ext}(\mathbf{r}) + \int d^3r\, n(\mathbf{r}) V_H(\mathbf{r})}_{\textbf{Classical Electrostatics}} + \underset{\textbf{Correlations}}{E_{xc}[n]}$$

$E=E[n(\mathbf{r})]$ where the density $n(\mathbf{r})$ is the basic variable. In Density-Functional Theory, $E=E[n(\mathbf{r})]$ --- functional of n alone rather than the (vastly more complicated) $E=E[\{\psi_i\}]$.

They proved that $n(\mathbf{r})$ is determined by the variational principle:
$n(\mathbf{r})$ is the density where $E \rightarrow \min$.

The proof is implicit $T[n]$ and $E_{xc}[n]$ are universal but unknown
$T[n]$ is very pathological … it must be to describe e.g. core levels.

⇒ Tractable theory: Kohn-Sham ansatz: Assume $T[n]=T$ for a non-interacting system that generates n

Kohn-Sham Ansatz and the Variational Principle

$$E_{KS} = T_s[n] + E_{\text{pot}}[n], \quad E_{\text{pot}}[n] = \int d\mathbf{r}\, V_{\text{ext}}(\mathbf{r}) n(\mathbf{r}) + E_H[n] + E_{xc}[n]$$

Noninteracting ansatz provides a means for functional for T_{KS} .
Assume a noninteracting auxiliary system exists which
generates the same density. Its K.E. is known:

$$H_{KS}^{\text{eff}} \psi_i = -\frac{1}{2}\nabla^2 \psi_i + V^{\text{eff}}(\mathbf{r})\psi_i = \varepsilon_i \psi_i, \qquad V^{\text{eff}}(\mathbf{r}) = \frac{\delta E_{\text{pot}}[n(\mathbf{r})]}{\delta n}$$

Multiply by ψ^* and integrate $\langle \psi_i | -\frac{1}{2}\nabla^2 + V^{\text{eff}} | \psi_i \rangle = \langle \psi_i | \varepsilon_i | \psi_i \rangle = \varepsilon_i$

Sum over states i and note $T_{KS} = \sum_i \langle \psi_i | -\frac{1}{2}\nabla^2 | \psi_i \rangle$

Then

$$T_{KS} = \sum_i \varepsilon_i - \int d\mathbf{r}\, V^{\text{eff}}(\mathbf{r}) \underbrace{\sum_i \psi_i^*(\mathbf{r}) \psi_i(\mathbf{r})}_{\textbf{Density } n(\mathbf{r})} = E_{bs} - \int d\mathbf{r}\, V^{\text{in}}(\mathbf{r}) n^{\text{out}}(\mathbf{r})$$

Thus we get at T_{KS} through a back door.

Kohn-Sham Ansatz in practice

DFT in practice: Kohn-Sham energy functional

$$E_{KS} = T_s[n] + E_{\text{pot}}[n], \quad E_{\text{pot}}[n] = \int d\mathbf{r} V_{\text{ext}}(\mathbf{r}) n(\mathbf{r}) + E_H[n] + E_{xc}[n]$$

Ansatz:

Get T_{KS} from orbitals ψ_i through H^{eff} where

$$H^{\text{eff}}(\mathbf{r}) = \frac{\delta E_{KS}[n(\mathbf{r})]}{\delta \psi}$$

$$H_{KS}^{\text{eff}} \psi_i = -\nabla^2 \psi_i + V^{\text{eff}}(\mathbf{r}) \psi_i = \varepsilon_i \psi_i, \qquad V^{\text{eff}}(\mathbf{r}) = \frac{\delta E_{\text{pot}}[n(\mathbf{r})]}{\delta n}$$

☑ Tractable functional for the total energy and forces

☑ An effective non-interacting Schrodinger equation, with eigenfunctions ψ_i and eigenvalues ε_i.

☑ A self-consistent electron density $n(\mathbf{r}) = \sum_i^{\text{occ}} |\psi_i(\mathbf{r})|^2$

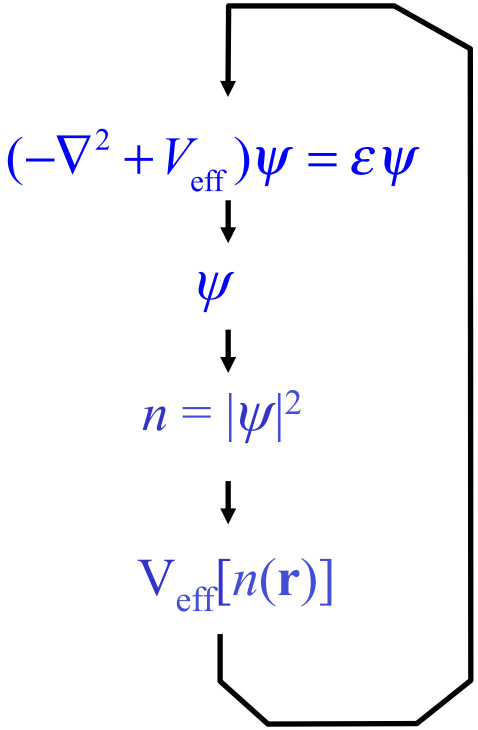
☒ Ansatz is not controlled or rigorously defined.

☒ ψ_i and ε_i are fictitious

Self-consistency

Chicken-and egg problem: The density $n(\mathbf{r})$ generates $V^{\mathrm{eff}}(\mathbf{r})$, while $V^{\mathrm{eff}}(\mathbf{r})$ generates $n(\mathbf{r})$ through $V^{\mathrm{eff}}(\mathbf{r}) \rightarrow \psi(\mathbf{r}) \rightarrow n(\mathbf{r})$.
Need one to get the other

Resolution: *guess* a trial $V^{\mathrm{eff}}(\mathbf{r})$, and iterate until the V^{eff} that *generates* ψ is the same as the V^{eff} *generated by* ψ.



$$(-\nabla^2 + V_{\mathrm{eff}})\psi = \varepsilon\psi$$

$$\psi$$

$$n = |\psi|^2$$

$$\mathrm{V}_{\mathrm{eff}}[n(\mathbf{r})]$$

The Harris-Foulkes Functional

The Kohn-Sham functional is then a functional of n^{out} :

$$E_{KS}[V^{\text{in}}] = E_{bs} - \int d\mathbf{r} V^{\text{in}}(\mathbf{r}) n^{\text{out}}(\mathbf{r}) + E_{\text{pot}}[n^{\text{out}}]$$

$$\boxed{E_{\text{KS}} = E_{\text{KS}}[V^{\text{in}}]}$$

Alternative functional where n^{in} is the independent variable.

$$E_{HF}[n^{\text{in}}] = E_{bs} - \int d\mathbf{r} V[n^{\text{in}}(\mathbf{r})] n^{\text{in}}(\mathbf{r}) + E_{\text{pot}}[n^{\text{in}}]$$

$$\boxed{E_{\text{HF}} = E_{\text{HF}}[n^{\text{in}}]}$$

Originally derived by Harris, but found independently and better explained by Foulkes and Haydock (PRB 39, 152520).

A Legendre transformation [n^{in} and V^{in} are conjugate variables]

$E[S,V] \rightarrow F[T,V] = E[S,V] - TS$, or $F[T,V] \rightarrow G[T,p] = F[T,V] + pV$

$E_{\text{HF}} = E_{\text{KS}}$ at self-consistency, but differ when $\Delta n \neq 0$.

E_{KS} is variational ($E_{\text{KS}}[V^{\text{in}}] > E_{\text{KS}}[V_{KS}]$) while E_{HF} is only stationary :

Usually $E_{\text{HF}} \rightarrow$ max while $E_{\text{KS}} \rightarrow$ min (they bound solution).

Typically E_{HF} is better: it is less sensitive to Δn.

Practical Implementations of the LDA

Single-particle orbitals ψ_i are expanded in some basis set. Many ways to do this.

Classification #1: The ψ_i expanded in plane waves (most common), or in short-range, atom-centered orbitals.

Questaal uses the latter.

Classification #2: orthogonalization to the core. *Either*:

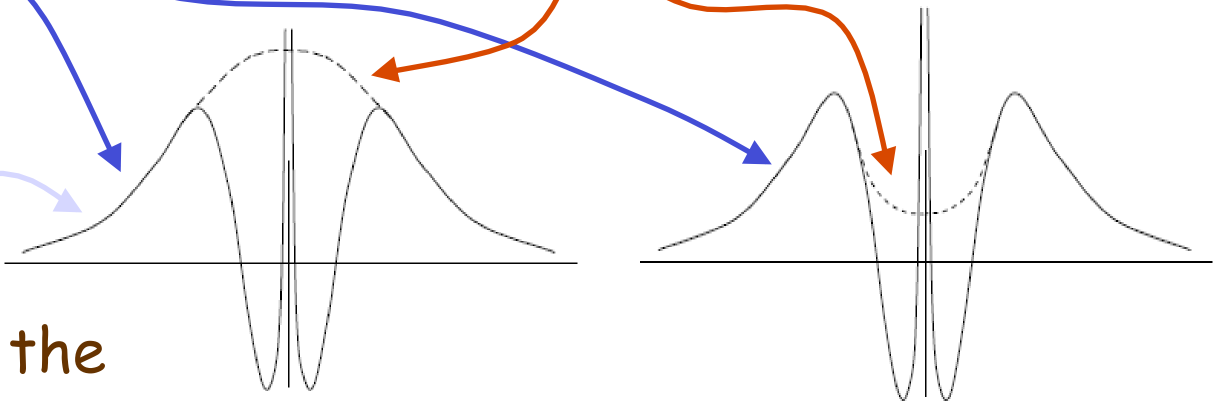
Replace the core by an effective (pseudo)potential, *or*

Keep all electrons: Augment the wave functions in spheres around each nucleus with numerical solutions of the radial Schrodinger equation. Originally formulated by Slater

Questaal is an all-electron method

Pseudopotentials

Add some artificial, or pseudo-potential V^{PS} to V^{LDA} ⇒ replace true wave function ψ with smooth analog of ψ^{PS} , with the same eigenvalues.



No core!

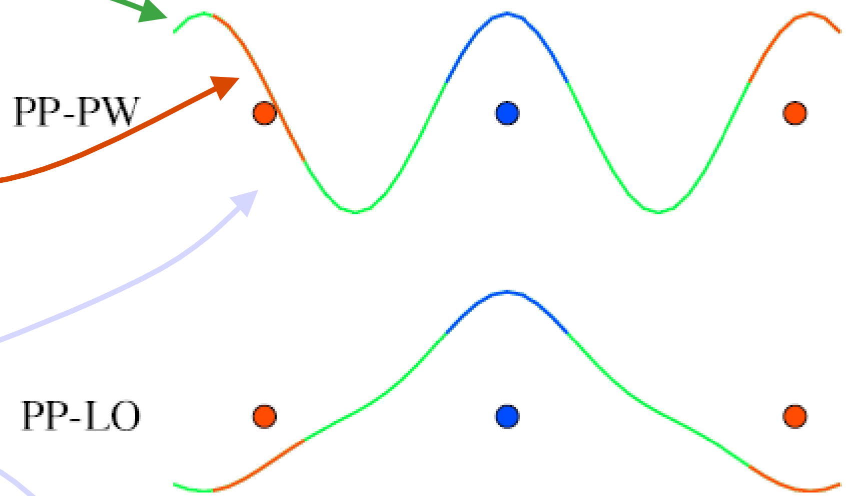
V^{PS} is not unique!

The envelope defines the basis.

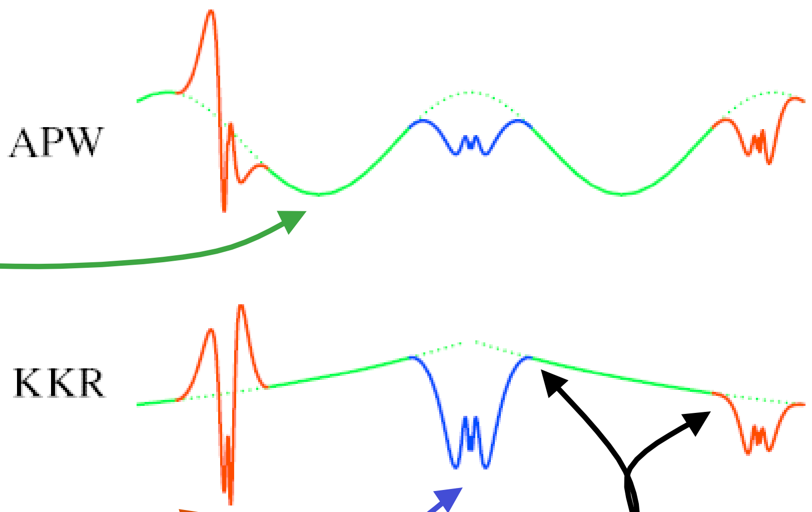
Shown here as green.

Blue, red show augmentation region : region where $V^{PS} \neq 0$.

Can be Plane Waves, or Atom-centered functions



Augmentation



All-electron basis sets begin with envelope functions :

Far from nuclei ψ is smooth and described by smooth envelopes. They can be either plane waves or local functions.

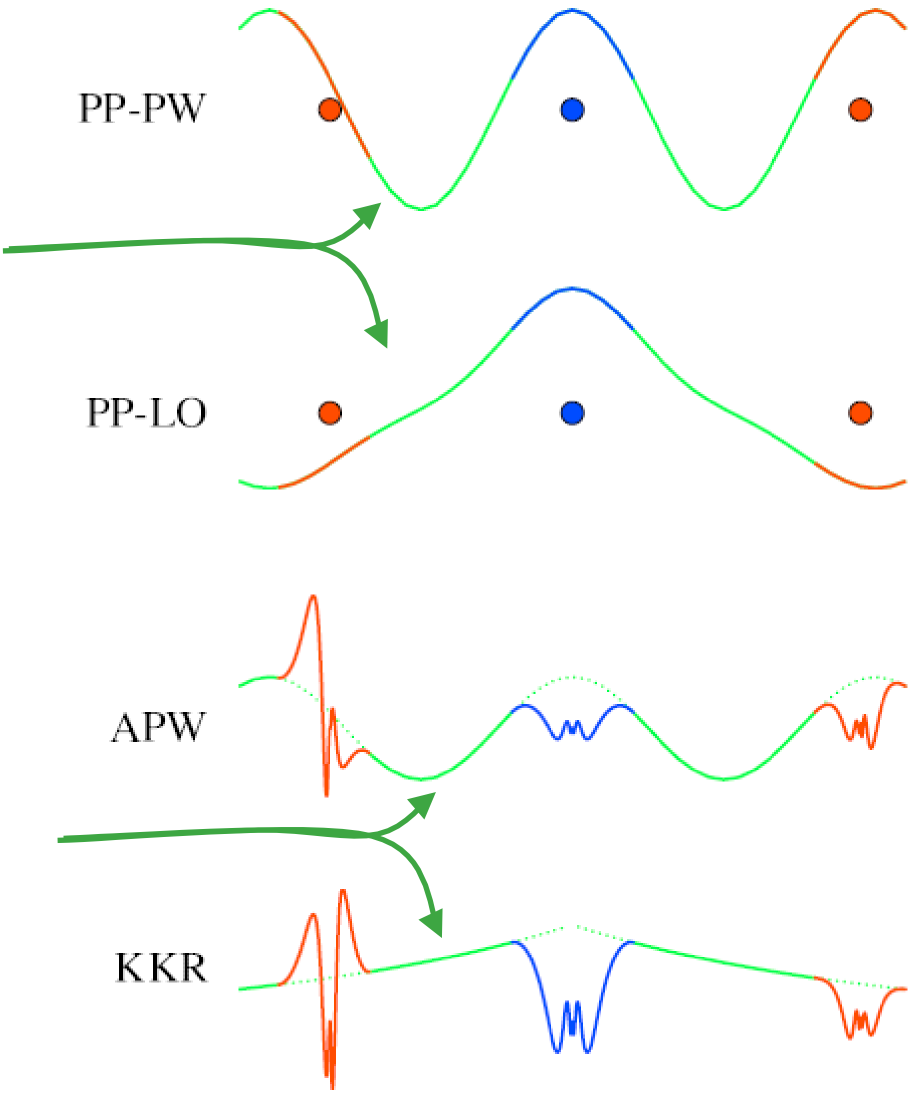
Valence ψ must be orthogonal to core states ⇒ many wiggles that envelope functions cannot well represent.

Solution: augment spheres around nucleus with numerical solutions of the Schrodinger equation (red, blue). Called partial waves.

At interface the augmented functions must match continuously and differentiably onto the envelopes.

Both value and slope must match ⇒ quantization condition (see later).

Four basic variants



4 types depicted in figure.

Envelope functions are green.
Blue, red show augmentation region

Pseudo functions:
☑ Very efficient
☒ Approximation can be an issue

All-electron :
☑ Most rigorous (e.g. semicores)
☒ More cumbersome

Plane waves:
☑ Readily made complete
☒ Not localized

Atom centered functions:
☑ Efficient, physical intuition
☒ Completeness can be an issue

Energy dependence

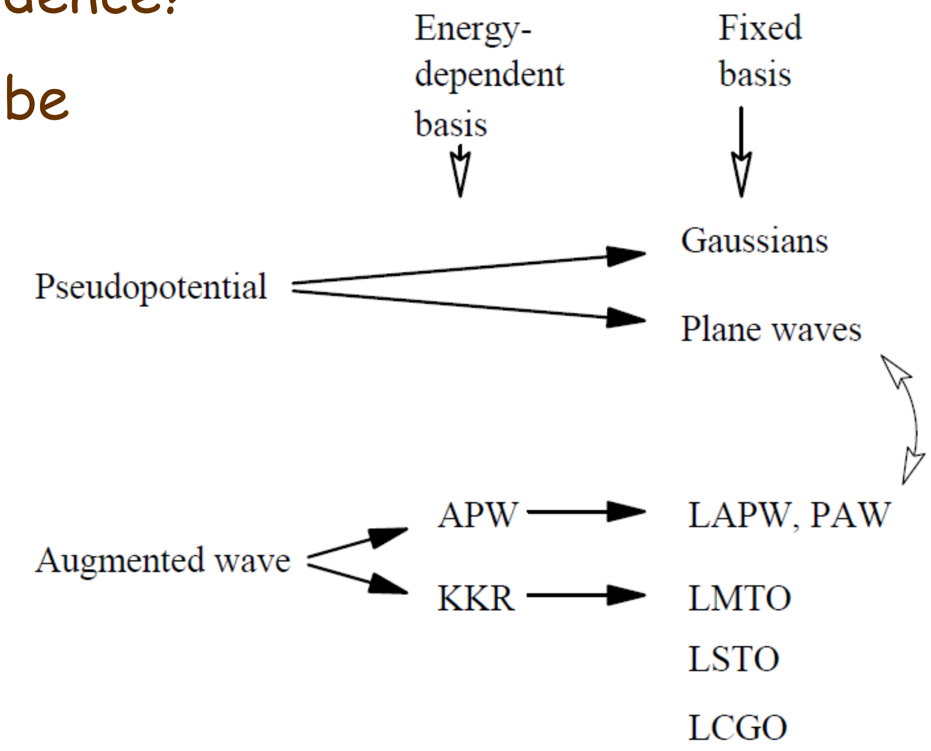
$\psi = \psi(E,\mathbf{r})$. What about E dependence?

Classification #3: the basis can be

(1) Fixed (PP methods) -- simplest

(2) potential-and-energy dependent (KKR,APW). Accurate but expensive. (Nonlinear eigenvalue problem; see later)

(3) potential dependent.
LAPW = linearized APW
LMTO = linearized KKR



Energy-dependent basis

Fixed basis

Pseudopotential

Gaussians

Plane waves

Augmented wave

APW

KKR

LAPW, PAW

LMTO

LSTO

LCGO

Linearization turns nonlinear secular matrix into linear algebraic eigenvalue problem, like fixed-basis case (later).

Modest loss of accuracy with large efficiency gains.

Muffin-tin potential and partial waves

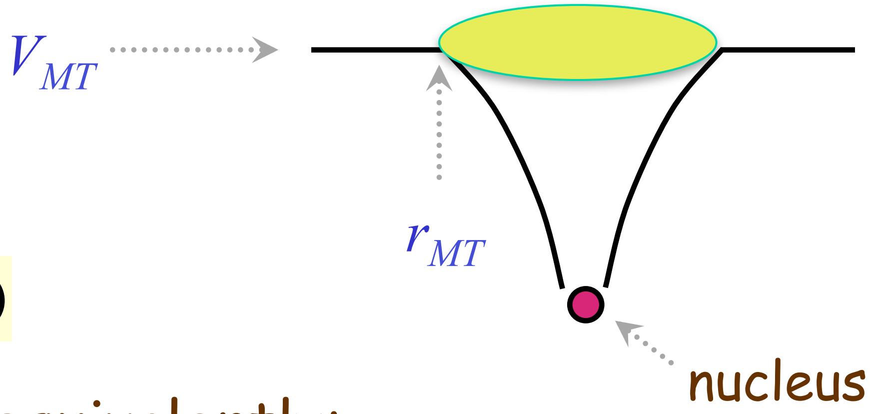
Cut and paste approach to Schrödinger equation

MT potential: spherically symmetric inside sphere (called "augmentation" sphere)
Constant in between

Construct basis function χ_j piecewise:

Consider the MT part. l and m are good quantum numbers; use L as a shorthand for compound (l,m) indices. The SE separates and:

$$\Phi_L(\varepsilon,\mathbf{r}) = Y_L(\hat{\mathbf{r}})\phi_l(\varepsilon,r)$$



ϕ_l satisfies $(-\nabla^2 + V(r))\phi_l = \varepsilon\phi_l$ or equivalently:

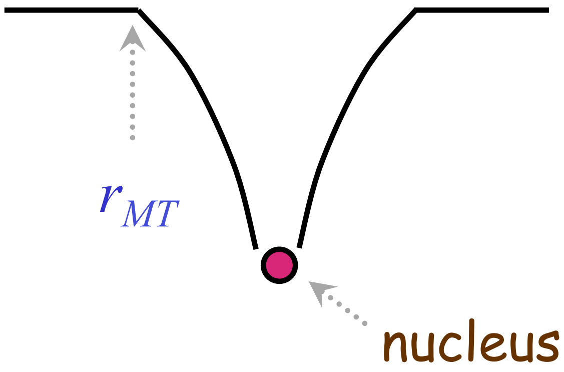
$$\left(-\frac{d^2}{dr^2} + V(r) + \frac{l(l+1)}{r^2} - \varepsilon\right) r\phi_l(\epsilon,r) = 0$$

The $\phi_l(\varepsilon,r)$ are called partial waves

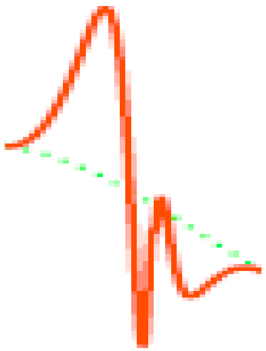
Numerics: logarithmic radial mesh

Inside r_{MT},

$$\left(-\frac{d^2}{dr^2}+V(r)+\frac{l(l+1)}{r^2}-\varepsilon\right)r\phi_l(\epsilon,r)=0$$



Solution must be orthogonal to cores ⇒ varies rapidly



Must be solved numerically on a radial mesh of points r_i:

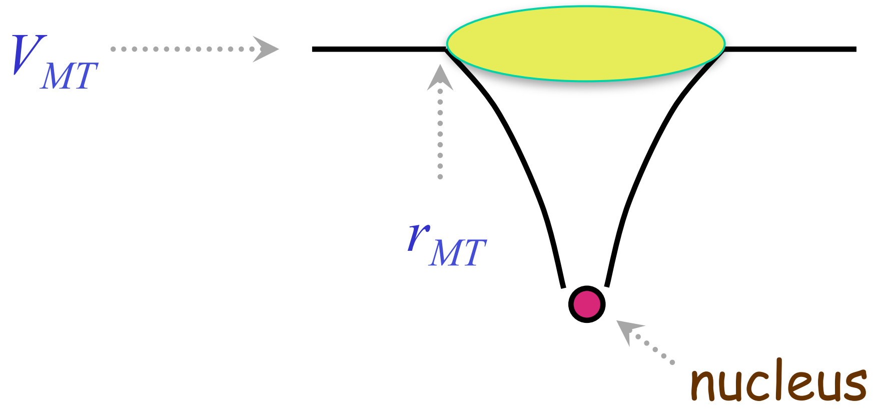
Customary: use one of:

logarithmic mesh $r_i = be^{ai}$ ⟵ **Simpler, more common**

shifted logarithmic mesh $r_i = b[e^{a(i-1)} - 1]$ ⟵ **Includes the origin**

Questaal code adopts a shifted logarithmic mesh

Partial Waves and Boundary conditions



Consider a single muffin-tin.

l and m are good quantum numbers.

Consider only the valence state (suppress index n) and use L as a shorthand for compound l+m indices. The partial wave is then

$$\Phi_L(\mathbf{r}) = Y_L(\hat{\mathbf{r}})\phi_l(r)$$

ϕ_l satisfies $(-\nabla^2 + V(r))\phi_l = \varepsilon\phi_l$ or equivalently a 1D eqn in u=$r\phi$

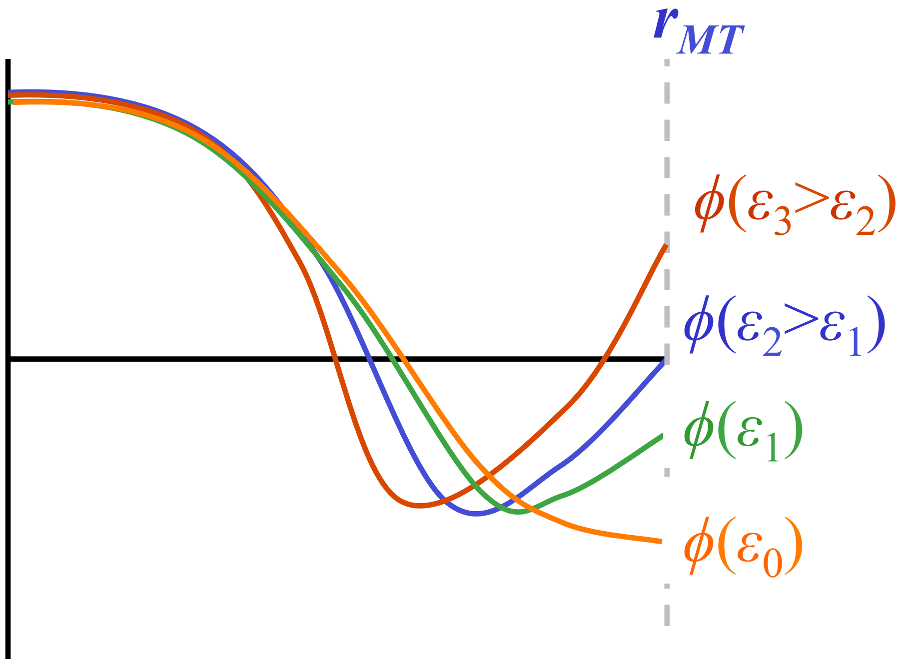
$$\left(-\frac{d^2}{dr^2} + V(r) + \frac{l(l+1)}{r^2} - \varepsilon\right)u_l(\text{Ú}, r) = 0, \qquad u_l(\text{Ú}, r) = r\phi_l(\text{Ú}, r)$$

$\underline{\Phi_l}$ is only a partial wave (incomplete solution) because we have yet to specify the boundary condition both as $r \rightarrow 0$ and at $r = r_{MT}$

2nd order differential equation ⇒ two solutions at any point.
Restriction: $\phi_l(r)$ regular as $r \rightarrow 0$ eliminates one b.c..
The boundary condition at r_{MT} will depend on the whole crystal.

Logarithmic Derivative I

Only one regular solution remains. Thus specifying ε completely specifies the remaining degree of freedom of the partial wave.



This information is neatly encapsulated in the logarithmic derivative at r_{MT}

$$D\{\phi_l(\varepsilon)\} = \left.\frac{r \cdot d\phi_l(\varepsilon)}{\phi \cdot dr}\right|_{r_{MT}} = \left.\frac{d\ln\phi_l(\varepsilon)}{d\ln r}\right|_{r_{MT}}$$

At some energy ε_0 the slope is negative

As ε increases, the curvature ($-\nabla^2$) increases.

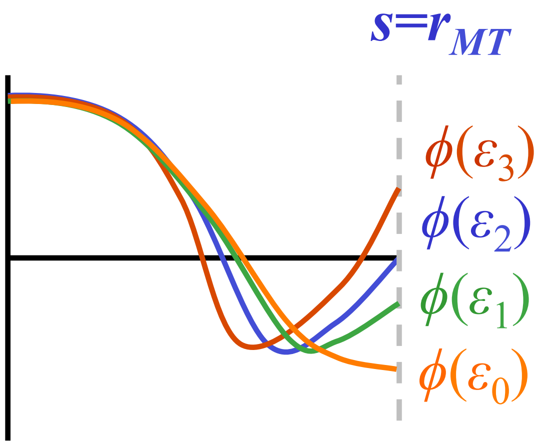
Between ε_0 and ε_1 slope $\phi'_l(r) \to 0$.

At ε_2, $\phi_l(r) \to 0$

Above ε_2 an extra node is picked up.

Logarithmic Derivative II

$$D\{\phi_l(\varepsilon)\} = \left.\frac{r\cdot d\phi_l(\varepsilon)}{\phi\cdot dr}\right|_{r_{MT}} = \left.\frac{d\ln\phi_l(\varepsilon)}{d\ln r}\right|_{r_{MT}}$$



$\varepsilon_0 : \phi_l < 0 \ \& \ \phi'_l < 0 \ \Rightarrow D > 0$

$\varepsilon_1 : \phi_l < 0 \ \& \ \phi'_l > 0 \ \Rightarrow D < 0$

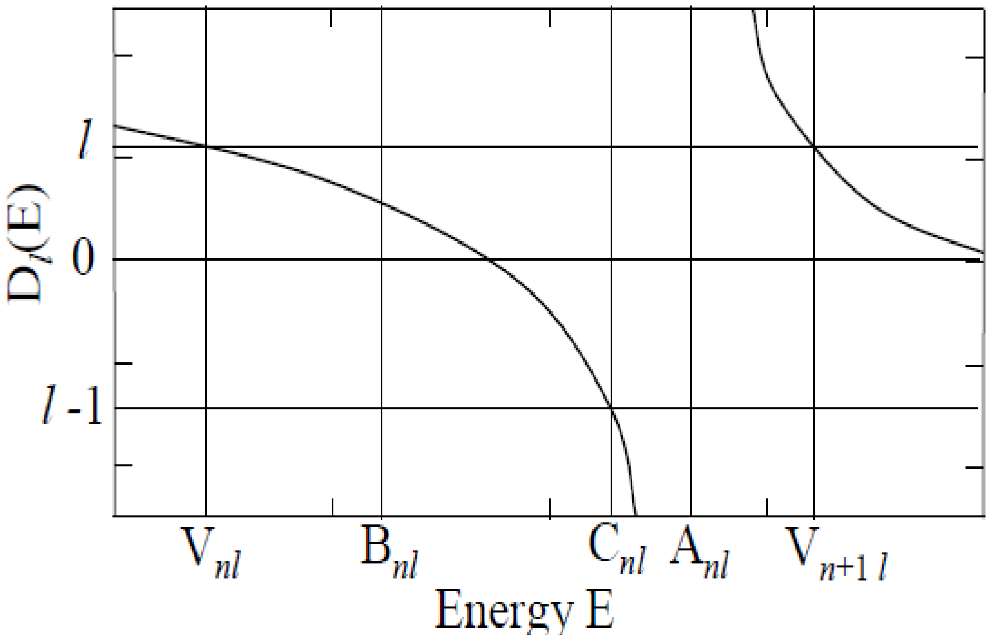
$\varepsilon_2 : \phi_l \to 0 \ \& \ \phi'_l > 0 \ \Rightarrow D \to \pm\infty$

$\varepsilon_3 : \phi_l > 0 \ \& \ \phi'_l > 0 \ \Rightarrow D > 0$

$D_l(\varepsilon)$ is an ever-decreasing, cotangent-like function.

PP methods construct V^{ps} by making $D_l^{\mathrm{peudo}} = D_l^{\mathrm{true}}$.

The “center of gravity” C_l (natural resonance) occurs near the point $D_l = -l$ (see later).

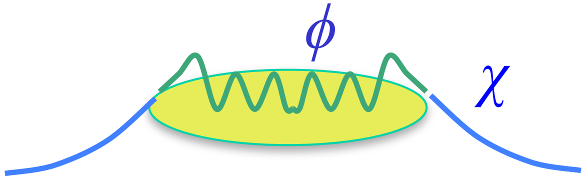


APW and KKR Methods

The $\phi_l(\varepsilon,r)$ join smoothly onto envelope functions in the interstitial.

Envelope function χ_j is "augmented" by $\phi_l(\varepsilon,r)$ in each augmentation sphere.

Envelopes can be any functions that are reasonably complete in the interstitial.



APW: Envelopes ⇒ plane waves $\chi_{\mathbf{G}} = e^{i(\mathbf{k}+\mathbf{G}).\mathbf{r}}$ as in the PW-PP method except $\chi_{\mathbf{G}}$ is augmented inside each MT sphere with partial waves.

KKR: If the potential really is a MT, it is solved exactly by

$$(\nabla^2 + \varepsilon - V_{MT})\,\chi(\varepsilon,\mathbf{r}) = 0, \quad r > r_{MT}$$

Helmholz equation: solns Hankels H and Bessels J

Family of Hankel functions $H_{\mathbf{R}L}(\mathbf{r})=H_L(\mathbf{r}-\mathbf{R})$ at each site $\mathbf{R}$: makes a nearly exact, minimal basis for the muffin-tin potential.

This is the KKR Method.

Spherical Hankel and Bessel Functions

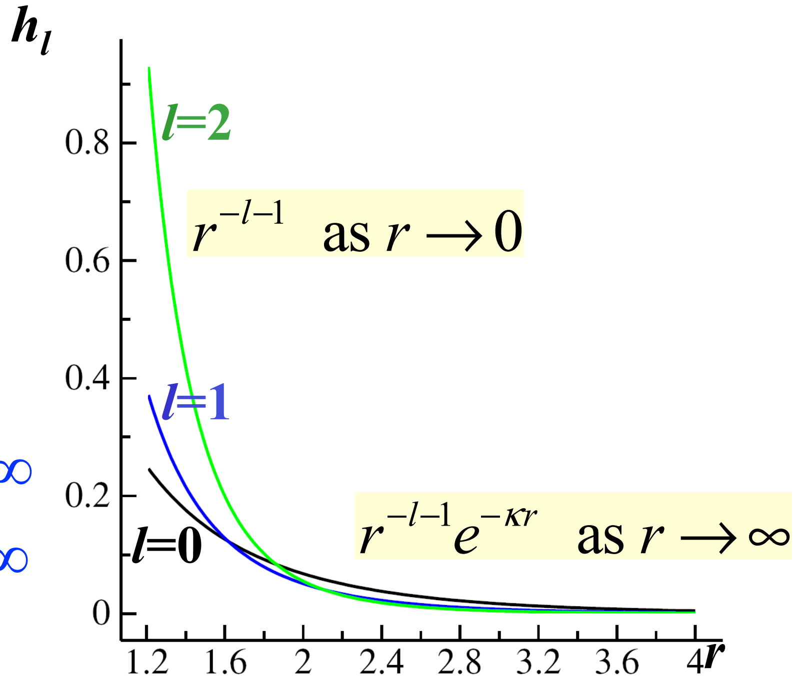
Spherical Hankel and Bessel functions satisfy this equation:

$$(\nabla^2+\kappa^2)\,\{H_L,J_L\}(\kappa^2,\mathbf{r})=0,\qquad r\neq 0$$

$$\{H_L,J_L\}(\kappa,\mathbf{r})=\{h_l,j_l\}(\kappa^2,r)Y_L(\hat{\mathbf{r}})$$

Radial part

Factor out angular dependence



r^{-l-1} as $r\to 0$

$r^{-l-1}e^{-\kappa r}$ as $r\to\infty$

Hankels: singular as $r\to 0$, regular as $r\to\infty$

Bessels: regular as $r\to 0$, singular as $r\to\infty$

$$h_l(\kappa^2,r)\to r^{-l-1}e^{-\kappa r}\quad \text{as } r\to\infty$$

$$j_l(\kappa^2,r)\to r^{l}e^{+\kappa r}\quad \text{as } r\to\infty$$

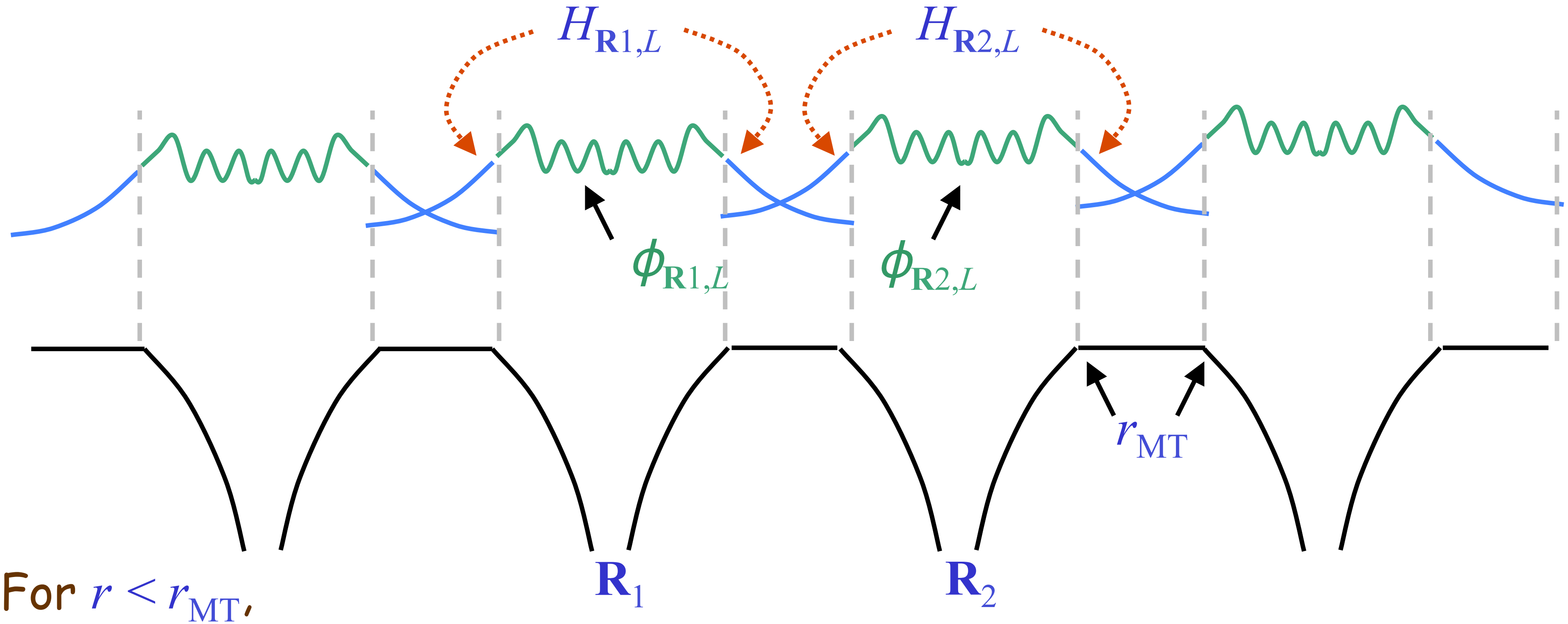
Hankels can be expanded as a linear combination of Bessels around a remote site. Called a one-center expansion:

$$H_L(\kappa^2,\mathbf{r}-\mathbf{R})=\sum_{L'}S(\mathbf{R}-\mathbf{R}',\kappa)J_{L'}(\kappa^2,\mathbf{r})$$

Structure constants

KKR method in a nutshell

In the interstitial, solutions are linear combinations of atom-centered Hankel functions $H_{\mathbf{R}L}$ centered at each nucleus $\mathbf{R}$.



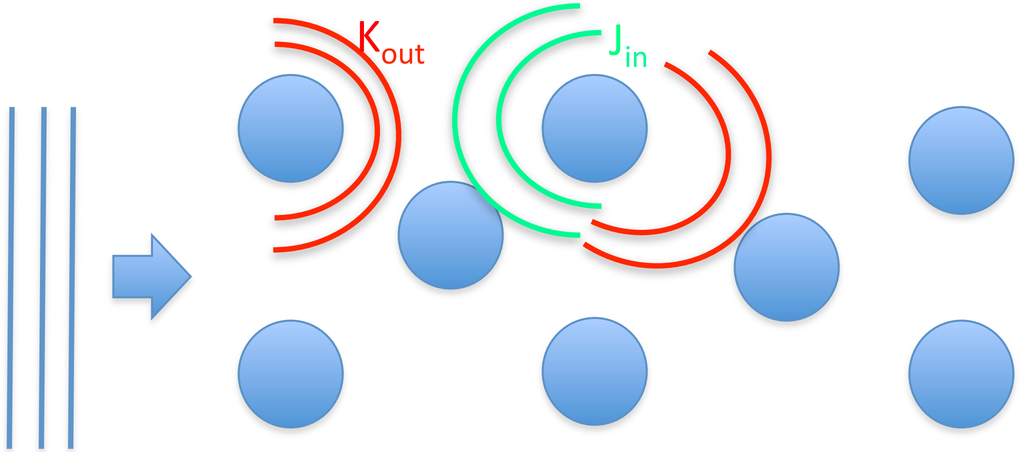
For $r < r_{\mathrm{MT}}$,

$$\Phi_{\mathbf{R}L}(\mathbf{r}) = \sum_{\mathbf{R}L} C_{\mathbf{R}L}\phi_{\mathbf{R}l}(\varepsilon, r) Y_L(\widehat{\mathbf{r}-\mathbf{R}}),$$

We have piecewise solutions of the SE, with kinks matching $H_{\mathbf{R}L}$ at r_{MT} .

To satisfy boundary conditions, kinks must disappear

Traditional view of Multiple Scattering theory (KKR)



Outgoing wave (H_{RL}) becomes incoming wave ($J_{R'L'}$) at neighboring sites.

H_{RL} scatters at each neighbor because of the MT potential there.

Sum of all scattered waves becomes incoming spherical wave on any given sphere.
Condition that they sustain each other without any original incoming plane wave.
Condition gives Multiple-Scattering equations

Sum over all paths

$$
\begin{aligned}
T_{ij} &= t_i\delta_{ij} + t_i G^0_{ij} t_j + t_i G^0_{ik} t_k G^0_{kj} t_j + \ldots \\
&= t_i\left[\delta_{ij} + G^0_{ij} t_j + G^0_{ik} t_k G^0_{kj} t_j + \ldots\right] \\
&= t_i\left[\delta_{ij} - G^0_{ij} t_j\right]^{-1} = \left[t_i^{-1}\delta_{ij} - G^0_{ij}\right]^{-1}
\end{aligned}
$$

t=tan (phaseshift)= t-matrix of each scatterer separately

G^0 is free electron propagator

KKR Maths

Define an energy dependent MTO as an “Augmented Hankel function”

$\Phi_{\mathbf{R}L}$ **: Partial wave centred at R**

Added to make χ **differentiable at R**

$$\begin{aligned}\chi_{RL}(\varepsilon,\mathbf{r}) &= N_{RL}(\varepsilon)\Phi_{RL}(\varepsilon,\mathbf{r}) + P_{RL}(\varepsilon)J_{RL}(\varepsilon,\mathbf{r}), & r<r_{MT}\\ &= H_{RL}(\varepsilon - V_{MT},\mathbf{r}), & r>r_{MT}\end{aligned}$$

$H_{\mathbf{R}L}$**: Hankel function centred at R**

$H_{\mathbf{R}L}$: exact soln of MT potential in interstitial

$\Phi_{\mathbf{R}L}$: exact soln of MT potential in sphere **R**

$\chi_{\mathbf{R}L}$ *would be* a solution if the $J_{\mathbf{R}L}$ “contaminates” the exact $\Phi_{\mathbf{R}L}$ ...

The eigenstate consists of a combination of $H_{\mathbf{R}'L'}$ centered at all sites. Inside **R**, : $H_{\mathbf{R}'L'}$ at $\mathbf{R}'\neq\mathbf{R}$ extend to **R** and because of the expansion theory, can be expressed in terms of the $J_{\mathbf{R}L}$

$$H_{R'L'}(\varepsilon,\mathbf{r}) = \sum_{L'} S_{RL,R'L'}(\varepsilon)J_{RL}(\varepsilon,\mathbf{r}) \quad \text{if } \mathbf{R}'\neq\mathbf{R}$$

KKR Maths II

$\Phi_{\mathbf{R}L}$ **: Partial wave centred at R**

Added to make χ differentiable at R

$$\begin{aligned}\chi_{RL}(\varepsilon,\mathbf{r}) &= N_{RL}(\varepsilon)\Phi_{RL}(\varepsilon,\mathbf{r}) + P_{RL}(\varepsilon)J_{RL}(\varepsilon,\mathbf{r}), \quad & r < r_{MT} \\ &= H_{RL}(\varepsilon,\mathbf{r}), & r > r_{MT}\end{aligned}$$

$H_{\mathbf{R}L}$**: Hankel function centred at R**

Idea : choose linear combinations $\Sigma_{\mathbf{R}'L'}[z_{\mathbf{R}'L'}H_{\mathbf{R}'L'}(\varepsilon,\mathbf{r})]$

so that the tails from neighbors $\mathbf{R}' \neq \mathbf{R}$ cancel the $J_{\mathbf{R}L}$ piece at $\mathbf{R}$ added to make $\chi_{\mathbf{R}L}$ continuous.

Do this by defining the

"Potential function" $P\{\phi_l(\varepsilon)\} = W\{\phi_l(\varepsilon), h_l\} / W\{\phi_l(\varepsilon), j_l\}$

Wronskian at r_{MT}

P specifies how much J you must add to $\chi_{\mathbf{R}L}$ to make it continuous at r_{MT}.

Properties of KKR

The “kink cancellation” condition can be written in this form:

$$|\mathbf{P}_{\mathbf{R}L}(\varepsilon) - \mathbf{S}_{\mathbf{R}L,\mathbf{R}'L'}(\varepsilon)| = 0$$

Potential function = amount of J in the head

Structure Matrix = amount of J from neighbors

$\mathbf{P}_{\mathbf{R}L}$: parameterizes information about scattering at r_{MT}.

All information about $V(\mathbf{r})$ at site $\mathbf{R}$ is folded into $P_{\mathbf{R}L}$

$P_{\mathbf{R}L}$ knows nothing about structure.

$S_{\mathbf{R}'L',\mathbf{R}L}$ contains information about structure

It knows nothing about potential.

KKR is elegant. Optimally compact for MT.
(MT potential ≅ true potential in close-packed structures).

But … solution involves a tedious energy search, with energy-dependent orbitals ⇒ inefficient, yields only G, not ψ.

Linear Methods in Band Theory

Problem …

APW & KKR ⇒ energy dependent hamiltonians.

Resolved by linearization

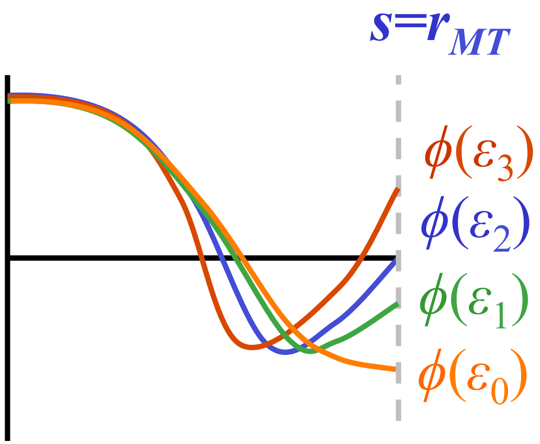
Ole Krogh Andersen

Nearly all modern electronic structure methods use linearization in some way

6,500 citations

- Linear methods in band theory O. K. Andersen Phys. Rev. B 12, 3060 (1975)
- Explicit, First-Principles Tight-Binding Theory, O. K. Andersen and O. Jepsen Phys. Rev. Lett. 53, 2571 (1984). See also Phys. Rev. B 34, 5253 (1986)
- Muffin-tin orbitals of arbitrary order, O. K. Andersen and T. Saha-Dasgupta Phys. Rev. B 62, R16219 (2000) and also chapter in Electronic Structure and Physical Properties of Solids. The Use of the LMTO Method. Easiest-to read but predecessor of NMTO: Tank & Arcangeli, Phys. Stat. Sol. (b) 217, 89

Linearization



Key observation: partial wave $\phi_l(\varepsilon,r)$ in sphere varies smoothly and slowly with energy

$$\phi_l(\varepsilon,r)=\phi_l(\varepsilon_\nu,r)+(\varepsilon-\varepsilon_\nu)\dot{\phi}_l(\varepsilon,r)+\ldots$$

Energy dependence parameterized by log derivative function

$$D\{\phi_l(\varepsilon)\}=d\ln\phi_l(\varepsilon)/d\ln r$$

Or by potential function (they are related)

$$P\{\phi_l(\varepsilon)\}=W\{\phi_l(\varepsilon),h_l\}/W\{\phi_l(\varepsilon),j_l\}$$

Linearize $\phi_l(\varepsilon)$ kink-cancellation ⇒ linear algebraic eigenvalue problem

APW → LAPW; KKR → LMTO

Gold standard **Efficient**

Parameterization of D and P

It is not difficult to derive the connection between P and D , and their properties when $\phi_l(\varepsilon,r)$ is linearized.
Skip derivation here : a bit tedious.

A particularly useful relation is:

$$P(\varepsilon)=\frac{(\varepsilon-C)}{\gamma(\varepsilon-C)+\Delta}=\left(\frac{\Delta}{\varepsilon-C}+\gamma\right)^{-1}$$

Note simple pole structure

where the "potential parameters" are expressed in terms of Wronskians

$$\gamma=\frac{1}{P[D\{\dot{\phi}\}]}=\frac{W\{J,\dot{\phi}\}}{W\{K,\dot{\phi}\}} \qquad C=\varepsilon_{\nu}-\frac{W\{K,\phi\}}{W\{K,\dot{\phi}\}} \qquad \sqrt{\Delta}=-\left(\frac{2}{w}\right)^{1/2}W\{J,\phi\}$$

C = "band center" = eigenvalue of isolated MT
Δ = "band width" = width of band, in absence of hybridization with other channels
γ = "band distortion"

Most of the essential physics of the hamiltonian is contained in C, Δ and structure constants S.

LMTO: Energy Linearization of KKR

By linearizating $\phi_l(\varepsilon)$: $\phi_l(\varepsilon,r)=\phi_l(\varepsilon_\nu,r)+(\varepsilon-\varepsilon_\nu)\dot{\phi}_l(\varepsilon,r)+\ldots$

P can be parameterized by a simple pole structure

Band width

$$P^{-1}=\frac{\Delta}{\varepsilon-C}+\gamma$$

Band “distortion”

Band center-of-gravity

The KKR condition

$$0=|P-S|=\left|\left(\frac{\Delta}{\varepsilon-C}+\gamma\right)^{-1}-S\right|$$

can be written as

$$\left|\varepsilon-C-\sqrt{\Delta}S^{\gamma}\sqrt{\Delta}\right|=0,\qquad \left(S^{\gamma}\right)^{-1}=S^{-1}-\gamma$$

This is equivalent to the linear algebraic eigenvalue problem

$H\psi=\varepsilon\psi$ with $H=C+\sqrt{\Delta}S^{\gamma}\sqrt{\Delta}$

Implemented in ASA part of Questaal package
See https://www.questaal.org/docs/code/asaoverview/

LAPW method and MT approximations

The envelope functions can be any complete basis.

Alternative to atom-centered $H_{\mathbf{R}L}$: plane waves $\{e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}\}$

now $e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}$ is augmented inside each MT sphere with linear combinations of $\Phi_{\mathbf{R}L}$ and $J_{\mathbf{R}L}$ as before.

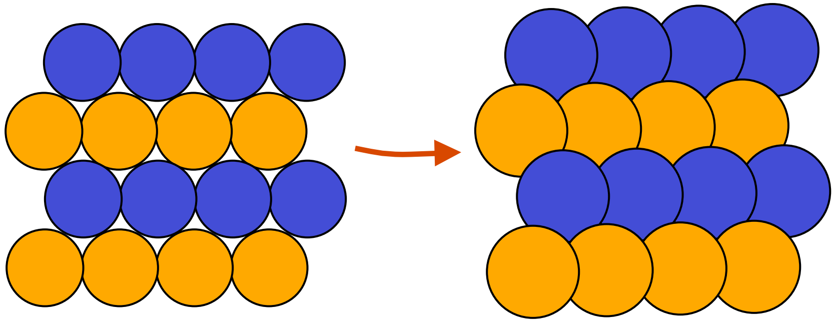
Similar "tail cancellation" condition ⇒ eigenfunctions.

But very expensive! Nonlinear eigenvalue problem. APW is rarely used today,

Linearization ⇒ LAPW method. Considered gold standard.

Traditionally KKR, APW, LMTO, LAPW (1980's) approximated the true potential by the MT form.

Enlarge spheres to fill space ⇒ "Atomic Spheres Approximation". Geometry violation better than MT.



All methods can be recast to eliminate the MT approximation and can solve the SE to high accuracy.

ASA codes, lm, lmgf, lmpg

The Questaal package has three kinds of ASA codes.

They are Noncollinear + with fully relativistic extensions

lm : band code computes self-consistent densities, energy bands for crystals (www.questaal.org/docs/code/asaoverview)

lmgf : similar purpose for crystals, using a Green's function technique (www.questaal.org/docs/code/lmgf/).
Additional functionalities particularly useful for JMRAM:

The coherent potential approximation for treating alloys and/or spin disorder

Linear response capabilities for computing magnetic exchange interactions

lmpg : a layer Green's function technique for transport through layered systems (www.questaal.org/docs/code/lmpg)

Attributes of the LMTO-ASA method

Positive attributes:

Very fast and efficient: can study many systems parametrically, e.g. response of a system to an external field.

The original LDA+U method, (nearly) the first noncollinear magnetism and magnetic exchange calculations, the first all-electron GW methods were built around it.

Connection with GF enables linear response calculations of e.g. magnetic exchange interactions, and CPA alloy theory

Ideally suited for, e.g. spintronics.

Drawbacks:

No forces

Accuracy is limited; results often must be checked

For many properties, other methods to do the same job better, or have functionality not in these methods.

Noncollinear Magnetism for large systems

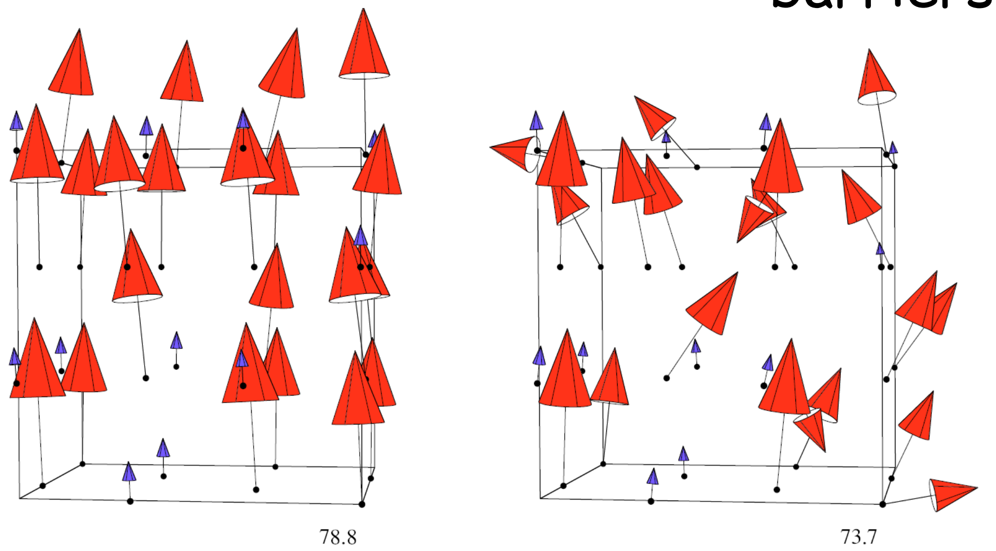
ASA band code lm … very versatile and efficient.

Early paper on
LDA calculations
of Schottky
barriers

VOLUME 65, NUMBER 21 PHYSICAL REVIEW LETTERS 19 NOVEMBER 1990

Electronic Structure of Ideal Metal/GaAs Contacts

Noncollinear magnetism-
good for spinstronics,
explained origins of
INVAR (Nature 400, 46).



First description of LDA-
based spin dynamics

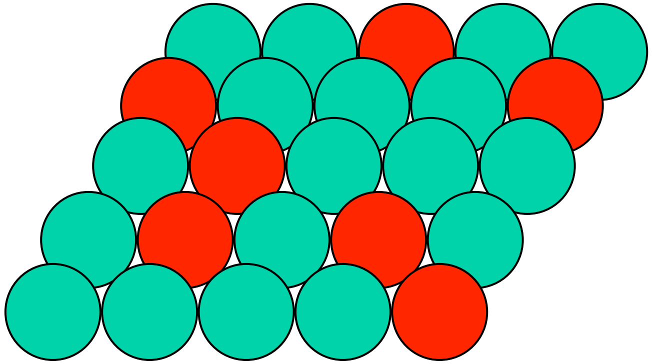
VOLUME 75, NUMBER 4 PHYSICAL REVIEW LETTERS 24 JULY 1995

***Ab Initio* Spin Dynamics in Magnets**

Coherent Potential Approximation, lmgf

The CPA provides an elegant way to model disorder on a lattice.

Different constituents of an alloy, e.g. Fe and Ni in permalloy (PY=$Fe_{0.2}Ni_{0.8}$) occupy sites randomly (more or less)



Crystal : the kink condition is $|P-S|=0$

Alloy : find a configurationally averaged "coherent potential function" $\mathcal{P}$ so the kink condition becomes $|\mathcal{P}-S|=0$

Binary alloy: atoms A and B occupy a site with probabilities c^A and $c^B=1-c^A$. Treat this site as an "impurity" embedded in an effective medium defined by the (as yet determined) coherent potential function $\mathcal{P}$

Coherent Potential Approximation II

From scattering theory, scattering from the impurity is given by the T-matrix. There is one for each atom $Q=1,2$:

$$t^Q = (P^Q - \mathcal{P})\left[1+(\mathcal{P}-S)^{-1}(P^Q-\mathcal{P})\right]^{-1} \qquad (1)$$

For there to be no net scattering on average (coherent potential), impose the condition

$$\sum_{Q=A,B} c^Q t^Q = 0 \qquad (2)$$

(1) and (2) constitute two simultaneous (matrix) equations for t and $\mathcal{P}$. They must be solved iteratively to self-consistency.

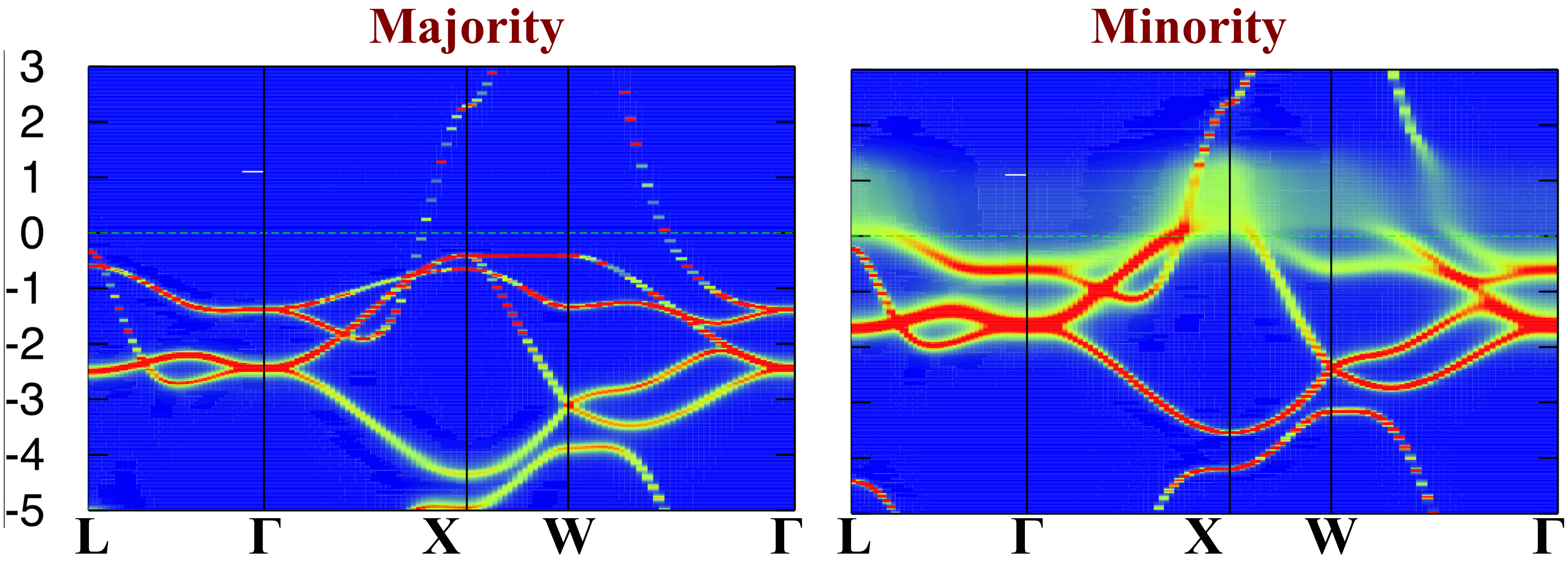
The CPA introduces an added complexity of an extra self-consistency step. $\mathcal{P}$ is now both complex and energy-dependent. Thus it cannot be converted to a linear algebraic eigenvalue problem. $\mathrm{Im}\,\mathcal{P}$ is the alloy contribution to the quasiparticle lifetime, and is a direct measure of the alloy scattering rate.

CPA Energy Band Structure of Permalloy (lmgf)

Alloy scattering : ⇒ k is not a good quantum number

In CPA, this gets reflected in a dynamical self-energy $\Sigma(\omega,\mathbf{k})$.

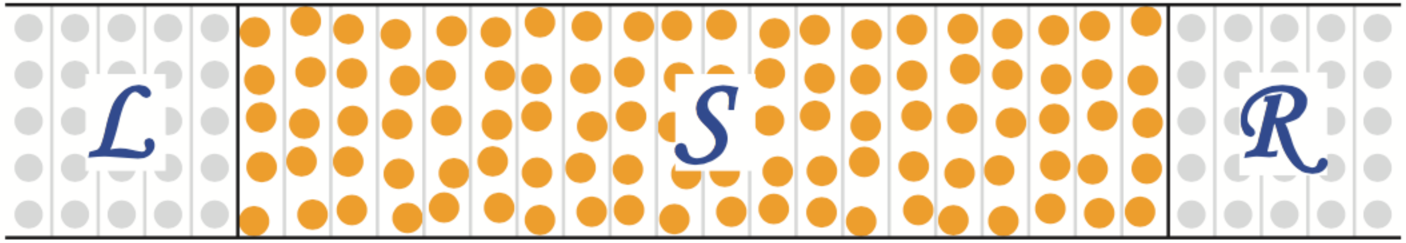
$\Sigma(\omega,\mathbf{k})$ has an imaginary component, which broadens the states.



Moderate scattering in majority channel, strong scattering in minority channel. Easily interpreted in terms of the potential parameters C and Δ.

Principal layer ASA-Green's function code lmpg

Layer geometry : Active (Scattering) region $\mathcal{S}$ cladded by semi-infinite leads $\mathcal{L}$ and $\mathcal{R}$.



Leads are assumed to be in equilibrium.

- Designed specifically for Landauer-Buttiker transport through layered systems :
- Periodic in 2 dimensions, not the third

lmpg has NEGF capability (non-equilibrium, Keldysh).
NEGF enables:
(1) beyond linear response
(2) inelastic scattering

Spin Flip Scattering at Metallic Interfaces (lmpg)


PRL **117,** 207204 (2016) PHYSICAL REVIEW LETTERS week ending 11 NOVEMBER 2016

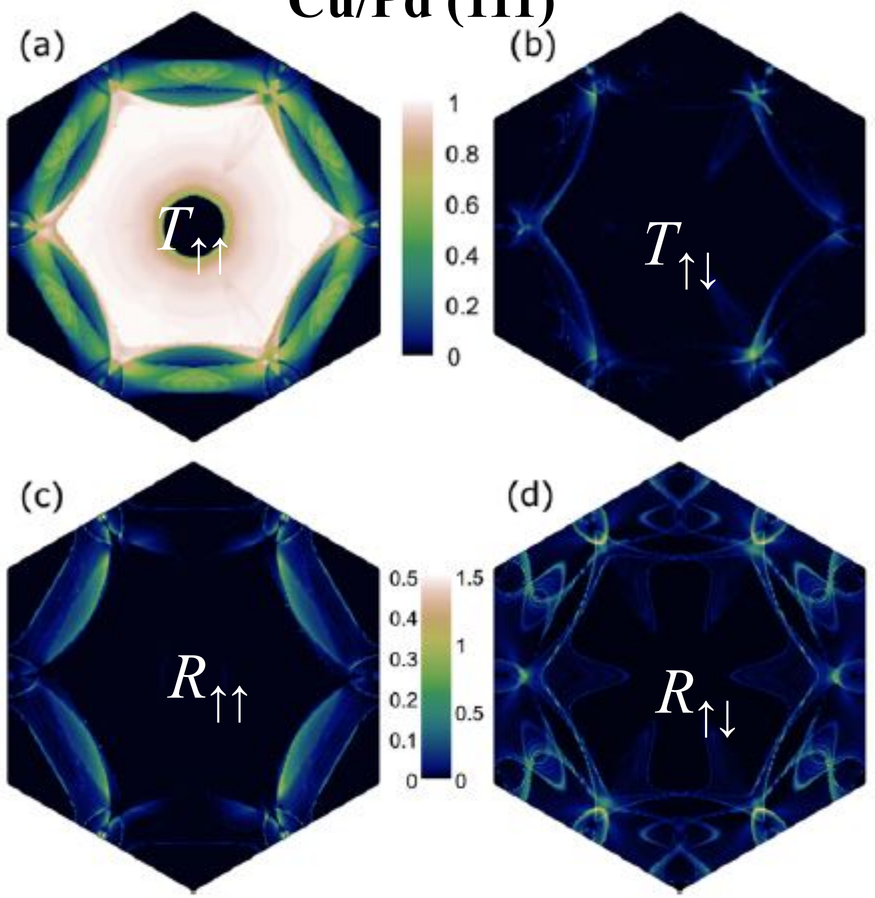
K. D. Belashchenko,[1] Alexey A. Kovalev,[1] and M. van Schilfgaarde[2]

Spin-flip scattering at interfaces contributes to spin relaxation in metallic multilayers

lmpg is designed to directly calculate spin-conserving and spin-flip transmission ($T_{\uparrow\uparrow}$ and $T_{\uparrow\downarrow}$) and reflectances ($R_{\uparrow\downarrow}$, $R_{\uparrow\downarrow}$)

In this Cu/Pd interface, the integrated spin flip probability was found to be approximately independent of surface intermixing.

Cu/Pd (111)



All-electron, full-potential methods

Full-potential methods usually do the following:

- Construct the basis set as though $V(\mathbf{r})$ were $V^{\mathrm{MT}}(\mathbf{r})$
- Exploit the variational principle to determine energy states.

Additional requirements to surmount the ASA approximation:

- More accurate --- need more complete envelope functions.
  And
- Ability to represent $n(\mathbf{r})$ and $V(\mathbf{r})$.

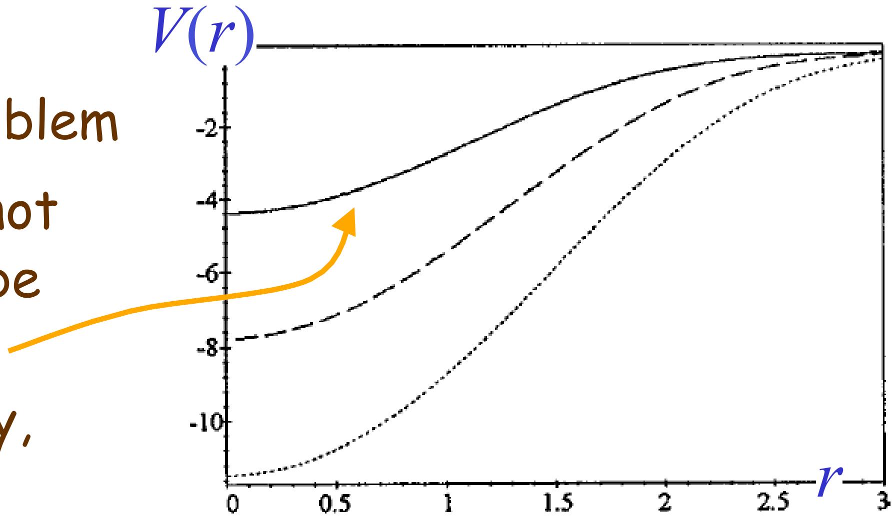
  Auxilliary basis for interstitial . We use plane waves.
- Matrix elements of the true potential: $\langle \chi_i \,|\, V(\mathbf{r}) - V^{\mathrm{MT}}(\mathbf{r}) \,| \chi_j \rangle$.
  Envelope functions should be smooth

Straightforward with LAPW; harder with LMTO

Smoothed Hankel Functions

Hankel functions:

- $\propto 1/r^{l+1}$ for small r … singularity a problem
- are “too stiff” : real potentials are not flat in interstitial. Functions would be better if they satisfied a SE that approached a constant asymptotically, more like what real potentials do



Fourier transform of $H_L(E;\mathbf{r})$:

$$H_L(E;\mathbf{q}) = \frac{-4\pi(iq)^l Y_L(i\mathbf{q})}{E-q^2}$$

FT of Gaussian w/ width r_s $\quad g(r_s;r) = e^{-(r/r_s)^2}$

$$g(r_s;\mathbf{q}) = e^{-4r_s^2q^2}$$

Define smoothed Hankel as product of $H_L(E,\mathbf{q})$ and $g(r_s,\mathbf{q})$

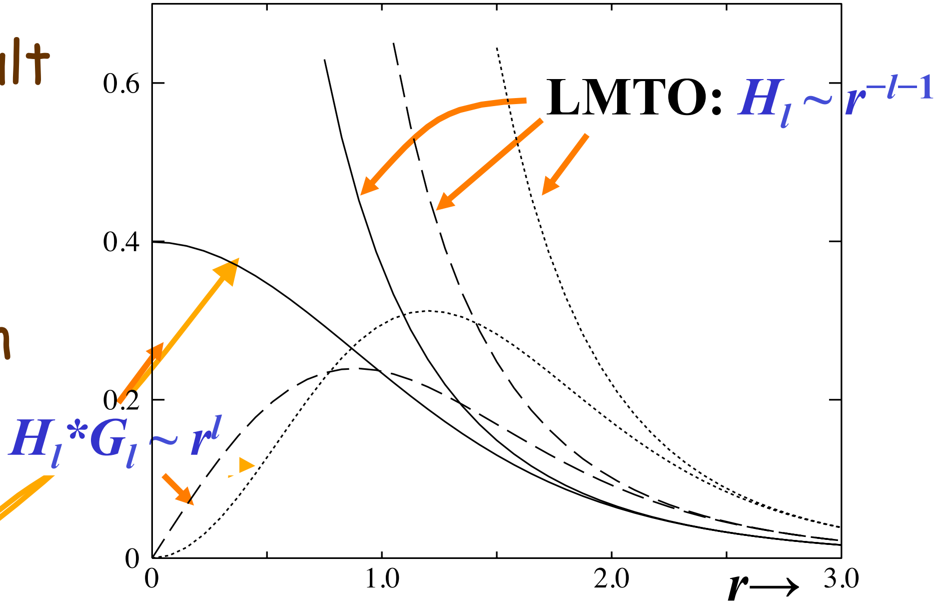
$$H_L^s(E,r_s;\mathbf{q}) = \frac{-4\pi(iq)^l Y_L(i\mathbf{q})e^{-4r_s^2q^2}}{E-q^2}$$

Smoothed Hankel Functions II

FT $H^s{}_L(E,r_s;\mathbf{q})$: to real space : result is a smoothed Hankel function.

Invented by M. Methfessel.

Properties thoroughly reviewed in J. Math. Phys. 39, 3393

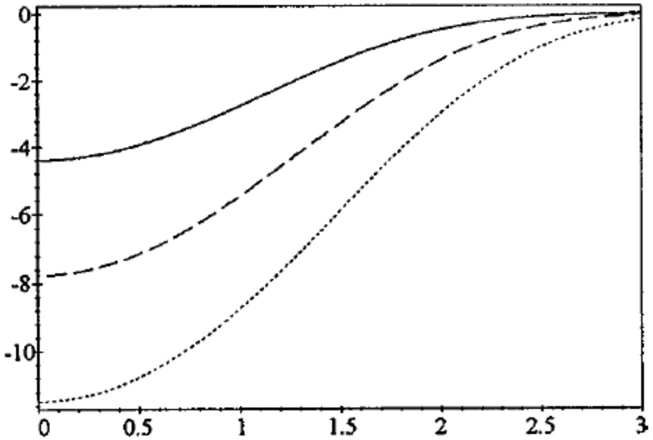


Key features:

1. Smooth everywhere
2. Regular at the origin : $H^s \sim r^l$ as compared to $H \sim r^{-l-1}$.
3. Satisfy a Schrodinger equation

$$\nabla^2 H_L = (-E + V(r))H_L,$$

$$V(r) = -4\pi \frac{G_L(r_s;r)}{H_L(E,r_s;r)}$$



Augmentation

Standard LAPW and LMTO methods must augment (replace the envelope function) with radial solutions of the SE to very high l … something like $l=6$ or 8.

Curious … because pseudopotential l-cutoffs are ~2. Yet there is a connection between augmentation and pseudopotentials. Why?

This code uses a unique form of augmentation.

The total density n is a superposition of 3 densities:

1. Interstitial, or smooth density n_i . Not augmented … extends everywhere in space (analogous to PP or PAW method)
2. True density inside augmentation sphere $\mathbf{R}$, $n_{1\mathbf{R}L}$
3. 1-center (local) representation of the interstitial density, $n_{2\mathbf{R}L}$

$$n(\mathbf{r}) = n_i(\mathbf{r}) + \sum_{\mathbf{R}L}^{L_{\max}} \left(n_{1\mathbf{R}L}(\mathbf{r}) - n_{2\mathbf{R}L}(\mathbf{r}) \right)$$

skip

Augmentation II

$$n(\mathbf{r}) = n_i(\mathbf{r}) + \sum_{\mathbf{R}L}^{L_{\max}} \left(n_{1\mathbf{R}L}(\mathbf{r}) - n_{2\mathbf{R}L}(\mathbf{r}) \right)$$

Properties of the "three-fold" representation of n:

1. A one-center expansion of n_i at site $\mathbf{R}$ becomes exactly to $\Sigma_L n_{2\mathbf{R}L}$ in the limit $L_{\max} \to \infty$ … thus n_i exactly cancels $n_{2\mathbf{R}}$ and the remaining density is the true density $n_{1\mathbf{R}}$. This establishes that the theory is exact if $L_{\max} \to \infty$.
2. In general $n_i \neq n_{2\mathbf{R}}$ inside the augmentation spheres. So for $L > L_{\max}$ the density is carried by n_i, and also n_i makes some nonzero contribution to n for $L < L_{\max}$.
3. For any $L_{\max}$, the value and slope of $n_{1\mathbf{R}}$ and $n_{1\mathbf{R}}$ exactly match at r_{MT}. Thus the net local density vanishes there, value and slope, and n is carried entirely by n_i.

skip

Augmentation III

The potential has a similar "three-fold" representation

$$V(\mathbf{r}) = V_i(\mathbf{r}) + \sum_{\mathbf{R}L}^{L_{\max}} \left(V_{1\mathbf{R}L}(\mathbf{r}) - V_{2\mathbf{R}L}(\mathbf{r}) \right)$$

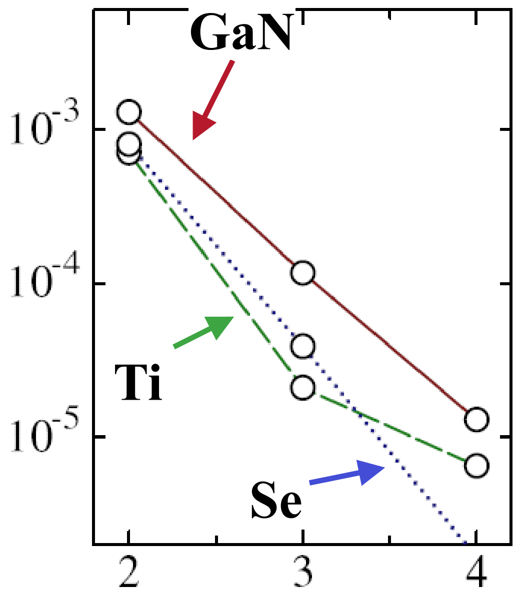
Each potential is computed (almost) independently of the others:

V_i is calculated from n_i only
$V_{1\mathbf{R}}$ is calculated from $n_{1\mathbf{R}}$ only
$V_{2\mathbf{R}}$ is calculated from $n_{2\mathbf{R}}$ only

No cross terms!

Why is this better?

- Cross terms are unwieldy
- L convergence much faster than standard LAPW
- Makes connection between PP and PAW methods, and all-electron methods.



Local orbitals

Recall energy linearization common to (nearly) all methods

$$\Phi_{\mathbf{R}L}(\varepsilon,\mathbf{r})=\phi_{\mathbf{R}l}(\varepsilon,\mathbf{r})Y_{\mathbf{R}L}(\hat{\mathbf{r}})$$

$$\phi_{\mathbf{R}l}(\varepsilon,\mathbf{r})\approx\phi_{\mathbf{R}l}(\varepsilon_v,\mathbf{r})+(\varepsilon-\varepsilon_v)\dot{\phi}_{\mathbf{R}l}(\varepsilon_v,\mathbf{r})+\dots$$

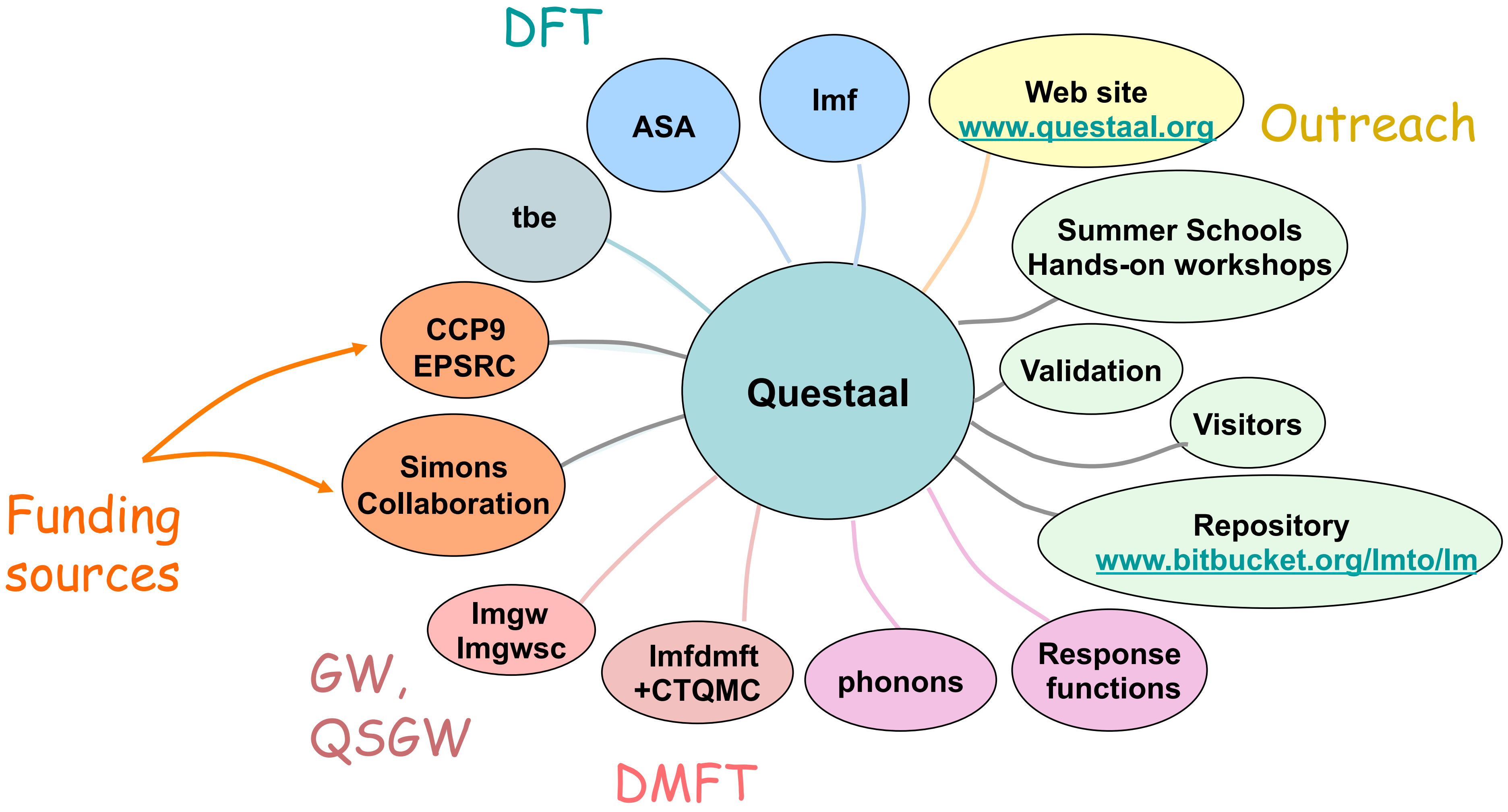
Standard: In the LMTO method (and LAPW) methods we add new augmentation orbitals $\phi_{\mathbf{R}L}=\phi_{\mathbf{R}L}(\varepsilon_v,r)$ and $\partial\phi_{\mathbf{R}L}/\partial\varepsilon$.   Procedure:

1. Integrate $\phi_{\mathbf{R}L}(\varepsilon_z,r)$ at some ε_z
2. Subtract $a\times\phi_{\mathbf{R}L} + b\times\partial\phi_{\mathbf{R}L}/\partial\varepsilon$ from $\phi_{\mathbf{R}L}(\varepsilon_z,r)$ to make both value, slope vanish at r_{MT} . The resulting "local orbital" is completely confined to the augmentation sphere.
3. Add this orbital to the basis set. Extends range of energy window which solves SE, and the cost of a larger basis.

CCP9 Flagship: Questaal suite

Questaal is the current CCP9 flagship (www.questaal.org)

It is an integrated electronic structure package that combines:



Questaal

DFT

- **ASA**
- **lmf**
- **tbe**

Outreach

- **Web site**
  www.questaal.org
- **Summer Schools**
  Hands-on workshops
- **Validation**
- **Visitors**
- **Repository**
  www.bitbucket.org/lmto/lm

Funding sources

- **CCP9**
  EPSRC
- **Simons Collaboration**

GW, QSGW

- **lmgw**
  lmgwsc

DMFT

- **lmfdmft**
  +CTQMC

- **phonons**
- **Response functions**

Descendant of the Stuttgart LMTO method

Problem …

APW & KKR ⇒ energy dependent hamiltonians.

Resolved by linearization

Ole Krogh Andersen

Nearly all modern electronic structure methods use linearization in some way

Linear methods in band theory O. K. Andersen Phys. Rev. B 12, 3060 (1975).

6,500 citations

Original LMTO-ASA was developed for the Stuttgart group.

Package formed the backbone of many developments :

- LDA+U (Aninismov)
- Exact exchange (Kotani)
- All-electron GW (Aryasetiawan)
- Electron-phonon interactions, superconductivity (Lichtenstein)

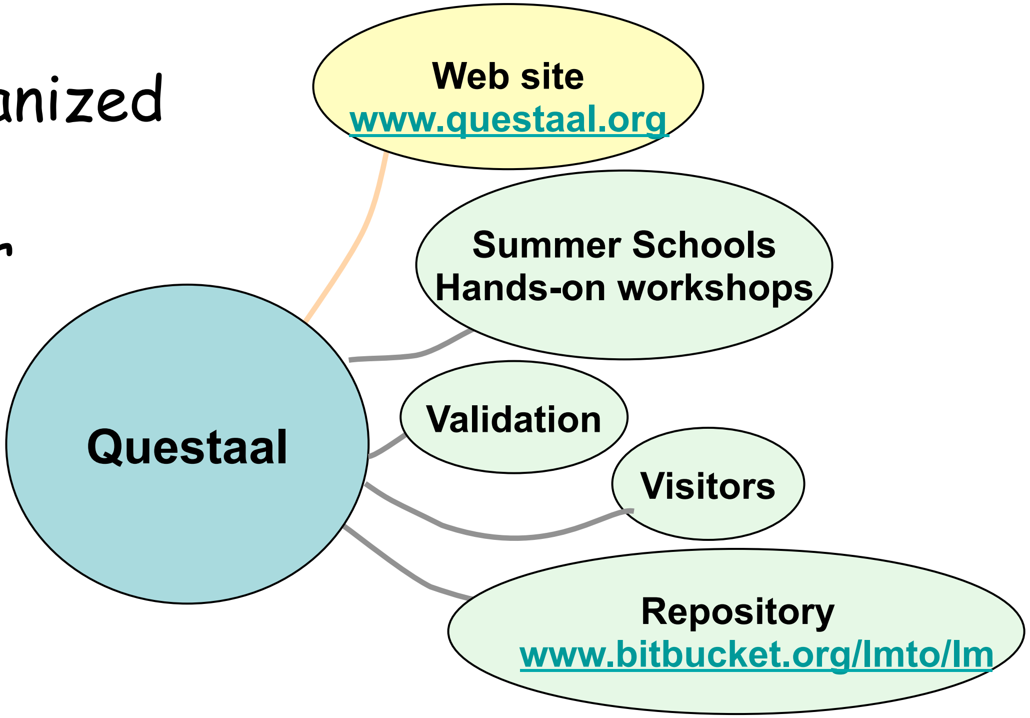
Full-potential version developed by Methfessel and MvS.

Full-Potential GW, QSGW developed by Kotani, Faleev, MvS.

Accessiblity

Make Questaal easily accessible w/ relatively low barriers to use

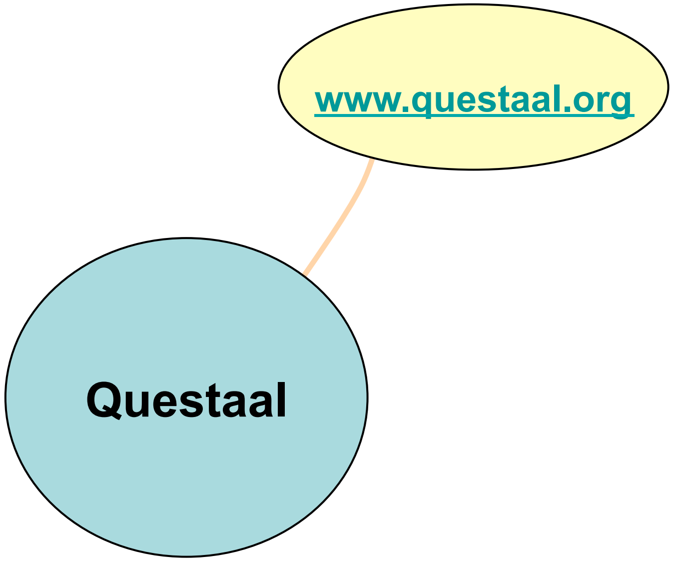
- Comprehensive and well organized web site with tutorials
- Logical and transparent user interface
- Hands on workshops
- Code validation
- Visitors
- Technical support



Goal: competent nonspecialists should be able to perform routine calculations.

You are our test case!

Web site



Web site built in the last year

- by undergrads Kantor and Christensen
- supervised by Pashov

Modern, with many nice features.

- Each main code documented.
- Basic tutorials for functionality
- Many missing links; still far from complete.

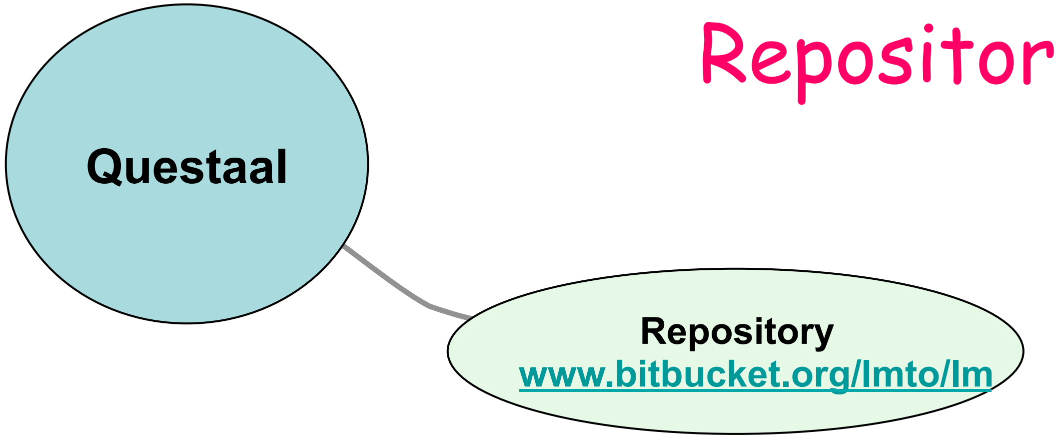
We hope you will help us identify problems with the web site!
If there are mistakes or you can't find something, let us know.

Web site built on Markdown (Kramdown) system

- Easily extended; intended for user community to add
- There is Style manual for uniform look and feel

We welcome contributions to the site (tutorials, new features)

Repository



Questaal

Repository
www.bitbucket.org/lmto/lm

Git repository for branching, history, version control

Modern, ninja-based build system

- Fast, parallel build
- One executable operates in both serial and MPI mode
- Validation: several hundred code checks

Some links to emerging hardware architectures

- GW code has interface to Xeon Phi

Validation exercise (Jackson)

Repository has ticketing system

Stack exchange planned

We have a (limited) visitor scheme

Functionality

DFT

Efficient basis set (Pashov)

Magnetic linear response, CPA

GW

Add diagrams (Gruening, Cunningham)

Improve efficiency (Pashov, Lueders)

Schemes to improve accuracy

Phonons

Phonons, electron-phonon interaction in GW (Bononi, Laricchia)

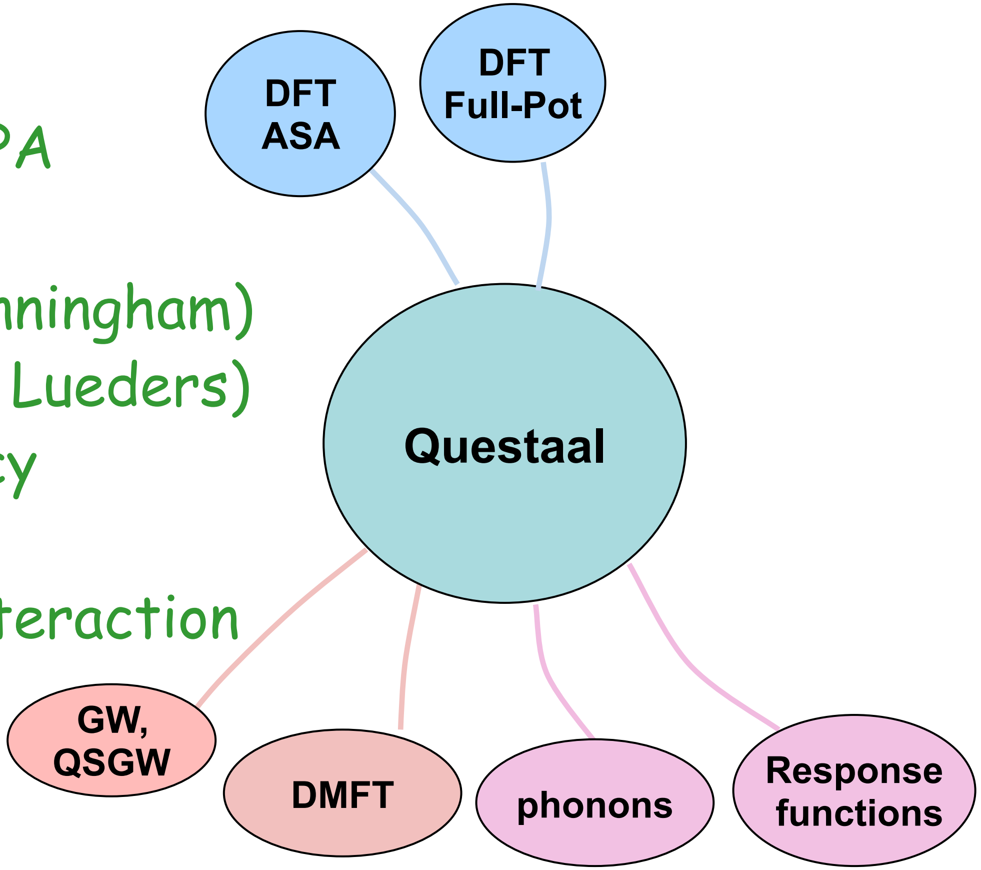
QSGW+DMFT

Acharya, in collabaration with Rutgers, Illinois, Paris

Response functions

Optical response, some magnetic susceptibility in MBPT

Magnetic susceptibility, some optical response, in DMFT



The GW approximation: Screened Hartree Fock

Hartree Fock: e^- senses an attractive potential V^x from antisymmetry that keeps e^- apart.

Write V^x in terms of Green's functions:

$$\Sigma_x = V_x(\mathbf{r}) = i\int G(\mathbf{r},\mathbf{r}')\frac{1}{|\mathbf{r}-\mathbf{r}'|}d^3\mathbf{r}' = iGv$$

GW: bare coulomb $v \rightarrow$ dynamically screened W:

$G =$ Green's Function, $W =$ screened coulomb interaction

$$V_{\text{bare}}(\mathbf{r},\mathbf{r}') = \frac{1}{|\mathbf{r}-\mathbf{r}'|} \rightarrow W(\mathbf{r},\mathbf{r}',\omega) = \varepsilon^{-1}v; \quad \Sigma = iGW$$

Dynamical screening is the essential difference between GW and Hartree Fock. It make both qualitative and quantitative changes to the electronic structure.

More rigorously: GW = the lowest order term in an exact expansion in W (many-body perturbation theory)

All-electron GW: Eigenfunction Products

The LDA works with products of basis function pairs, e.g. to make potential matrix elements $\langle \chi_i | V | \chi_j \rangle$ or $n(\mathbf{r})$.

GW is more complicated. It requires integrals of 4 basis functions with two-point quantities, e.g. matrix elements of coulomb interaction $v(\mathbf{r}, \mathbf{r}')=1/|\mathbf{r}-\mathbf{r}'|$ $\langle \Psi\Psi | v | \Psi\Psi \rangle$

Eigenfunction:

$$\psi_{\mathbf{k}n}(\mathbf{r}) = \sum_{Ru} \alpha^{\mathbf{k}n}_{Ru} \Phi^{\mathbf{k}}_{Ru}(\mathbf{r}) + \sum_{\mathbf{G}} \beta^{\mathbf{k}n}_{\mathbf{G}} P^{\mathbf{k}}_{\mathbf{G}}(\mathbf{r}),$$

$$P^{\mathbf{k}}_{\mathbf{G}}(\mathbf{r}) = \begin{cases} 0 & \text{if } \mathbf{r} \in \text{any MT} \\ \exp(i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}) & \text{otherwise} \end{cases}$$

Interstitial :

$$P^{\mathbf{k}_1}_{\mathbf{G}_1}(\mathbf{r}) \times P^{\mathbf{k}_2}_{\mathbf{G}_2}(\mathbf{r}) = P^{\mathbf{k}_1+\mathbf{k}_2}_{\mathbf{G}_1+\mathbf{G}_2}(\mathbf{r}) \qquad e^{-i(\mathbf{k}+\mathbf{G}')\cdot\mathbf{r}} \times e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}} = e^{-i(\mathbf{G}-\mathbf{G}')\cdot\mathbf{r}}$$

Augmentation

$$\Phi_L(\mathbf{r})\Phi_{L'}(\mathbf{r}) = \underbrace{\phi_l(r)\phi_l(r)}_{\text{Numerical solution}} \times \underbrace{Y_{L'}(\hat{\mathbf{r}})Y_{L'}(\hat{\mathbf{r}})}_{\text{Products of } Y_L}$$

$$Y_K(\hat{\mathbf{r}})Y_M(\hat{\mathbf{r}}) = \sum_L C_{KLM} Y_L(\hat{\mathbf{r}})$$

Product Basis

Interstitial: use $P_{\mathbf{G}_1}^{\mathbf{k}_1}(\mathbf{r}) \times P_{\mathbf{G}_2}^{\mathbf{k}_2}(\mathbf{r}) = P_{\mathbf{G}_1+\mathbf{G}_2}^{\mathbf{k}_1+\mathbf{k}_2}(\mathbf{r}), \quad P_{\mathbf{G}}^{\mathbf{k}} = e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}$

Augmentation: Construct complete set of product functions $B_N(\mathbf{r})$: $\{\varphi_{Ru}(\mathbf{r}) \times \varphi_{Ru'}(\mathbf{r})\}$ spanned by $B_N(\mathbf{r})$

R is a site index; u is a compound index which contains : L, label whether partial wave has phi, phidot, local orbital character.

In practice we must work with Bloch sums $B_I^{(\mathbf{k})}$ of the B_I.

Thus, the mixed basis completely spans the Hilbert space of basis function products $\{M_I^{\mathbf{k}}(\mathbf{r})\} \equiv \{P_{\mathbf{G}}^{\mathbf{k}}(\mathbf{r}), B_{RN}^{\mathbf{k}}(\mathbf{r})\}$

Example: Coulomb Matrix Elements

The bare coulomb operator can be expanded in the mixed basis:

$$v(\mathbf{r},\mathbf{r}') = \sum_{\mathbf{k},I,J} |\tilde{M}_I^{\mathbf{k}}(\mathbf{r})\rangle v_{IJ}(\mathbf{k}) \langle \tilde{M}_J^{\mathbf{k}}(\mathbf{r}')|$$

Similarly for all 2-point quantities, e.g. the dielectric function ε and screened coulomb interaction W. The mixed basis functions are not orthogonal so we must define

$$|\tilde{M}_I^{\mathbf{k}}\rangle \equiv \sum_{I'} |M_{I'}^{\mathbf{k}}\rangle (O^{\mathbf{k}})_{I'I}^{-1}, \quad O_{I'I}^{\mathbf{k}} = \langle M_{I'}^{\mathbf{k}} | M_I^{\mathbf{k}}\rangle.$$

Complete information to generate the GW self-energy are matrix elements like this.

Quasiparticle self-Consistent GW: Optimal G_0

Start with some trial V_{xc} (e.g. from LDA, or …). Defines G_0 :

$$H_0 = \frac{-1}{2m}\nabla^2 + V^{\mathrm{ext}}(\mathbf{r}) + V^{\mathrm{H}}(\mathbf{r}) + V^{\mathrm{xc}}(\mathbf{r},\mathbf{r}')$$

$$H_0\psi_i = E_i\psi_i \longrightarrow G_0(\mathbf{r},\mathbf{r}',\omega) = \sum_i \frac{\psi_i(\mathbf{r})\psi_i^*(\mathbf{r}')}{\omega - E_i}$$

GWA determines ΔV and thus H :

$$G_0 \xrightarrow{RPA} \varepsilon(iG_0G_0) \xrightarrow{GWA} \Sigma(\mathbf{r},\mathbf{r}',\omega) = iG_0W; \qquad \Delta V = \Sigma - V^{\mathrm{xc}}$$

Find a new V^{xc} that minimizes norm N, a measure of $\Delta V\, G_0$.

$$V^{\mathrm{xc}} = \frac{1}{2}\sum_{ij}\langle\psi_i|\mathrm{Re}\big(\Sigma(E_i)+\Sigma(E_j)\big)|\psi_j\rangle$$

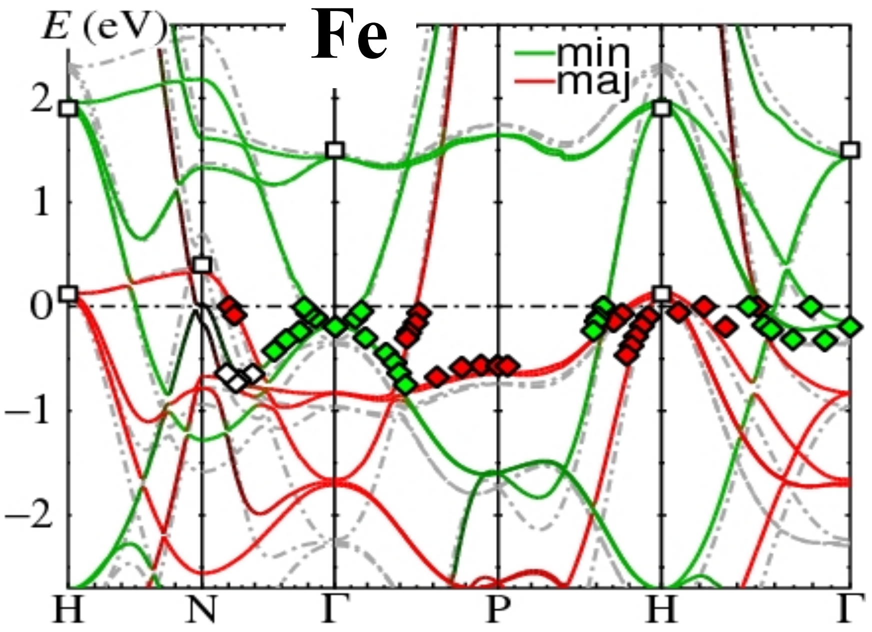
(approximate) result of min N

Iterate to self-consistency.

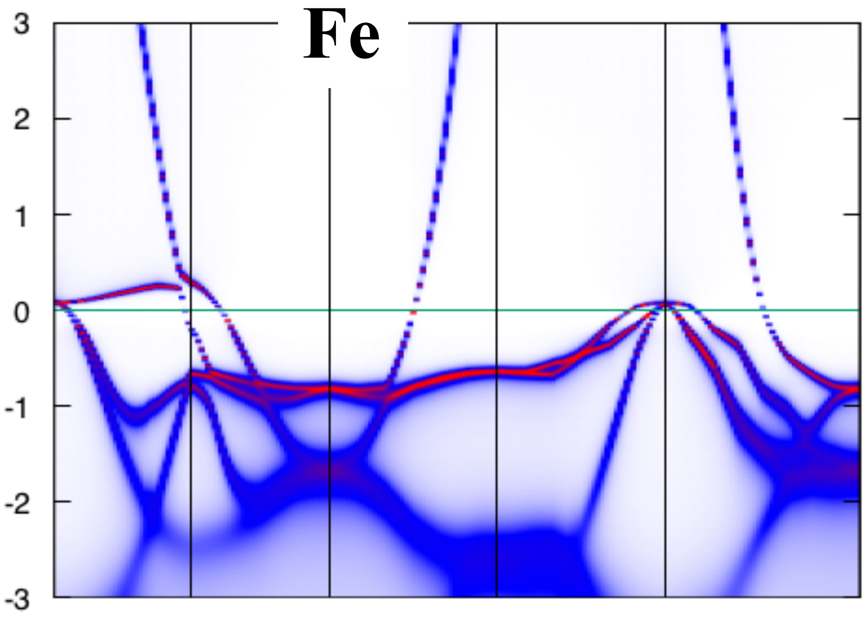
At self-consistency, E_i of G matches E_i of G_0 (real part).

Dual Nature of *QSGW*: both $G_0=(\omega-H_0)^{-1}$ and G

$E_i[G_0]$ = Re $E_i[G]$ ⇒ *QSGW* generates a nearly optimal noninteracting G_0 or H_0 for many kinds of materials classes …



G_0 majority+minority



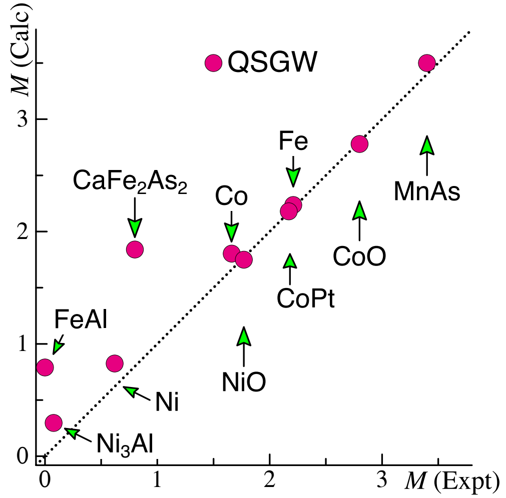
G majority only

But … as a many-body theory *QSGW* is limited …

Primary many-body effect is a plasmon. Superconductivity, Kondo physics, spin fluctuations … not captured.

Questaal's Implementation of QSGW+DMFT

Questaal has an interface to Kristjian Haule's CTQMC
(CTQMC = Continuous time Quantum Monte Carlo for DMFT)



Limited experience so far:

- Self-energies in QSGW+DMFT in a few compounds, e.g. La_2CuO_4.
- Special QSGW + "Magnetic DMFT" – Applied to Ni
- Ability to make two-particle quantities, e.g. spin susceptibilities.

Big challenges:

G_0 is not constructed from diagrams

What is local effective interaction U ?

Double counting