

VASP - Best Practices Workshop

Weine Olovsson

National Supercomputer Centre (NSC), Linköping University

NAIIS-ENCCS training, online 17-18th Apr 2024

VASP - Best Practices Workshop



<https://www.nsc.liu.se/>

NAISS

<https://www.naiss.se/>



EuroCC National Competence Centre Sweden

<https://enccs.se/>

National Supercomputer Centre (NSC)

NSC is part of:

- NAISS [National Academic Infrastructure for Supercomputing in Sweden](#)
- **li.u** LINKÖPING UNIVERSITY [liu.se](#)

NSC partners: [SAAB](#), [SMHI](#), [MET Norway](#)

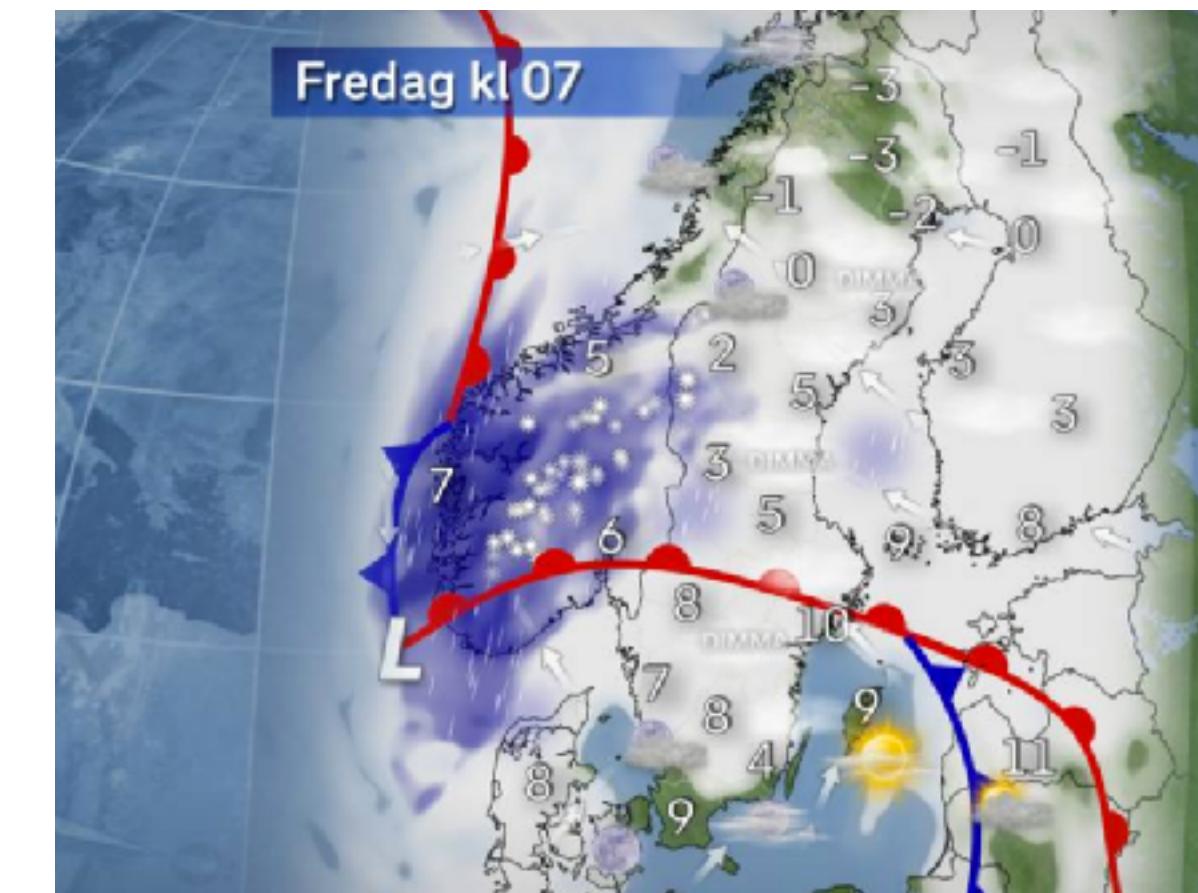


SAAB



SMHI

 Meteorologisk institutt



NSC academic clusters

Tetralith (2018 -) 1908 x 2 x 16 cores, Intel Xeon Gold 6130

NAISSL

Top500 no. 282 (74)

Sigma (2018 -) 110 x 2 x 16 cores, Intel Xeon Gold 6130



BerzeLiUs (2021 -) Nvidia DGX SuperPOD, 60 x 8 A100 GPUs

- increased with 34 nodes in 2023



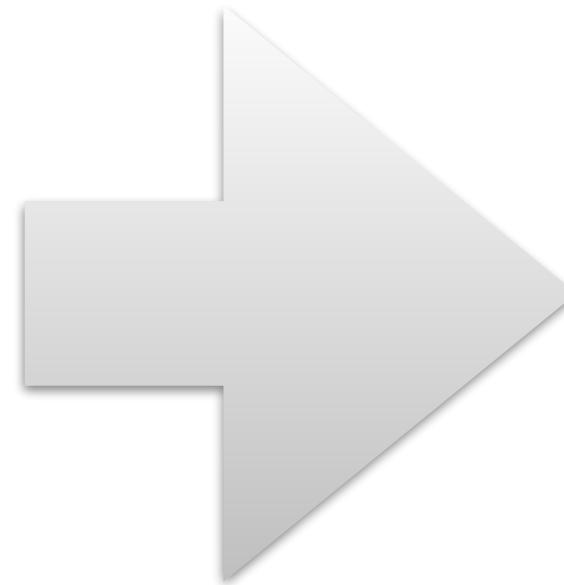
*Knut and Alice
Wallenberg
Foundation*

Top500 no. 151 & 178 (82)

About myself

- PhD in Physics 2005 @Uppsala Univ.
- PostDoc @Kyoto Univ. 4y, @Leoben Univ. 1y
- Application Expert @NSC, 2011 (50%), 2016 (90%)
- 10% theoretical spectroscopy @IFM, LiU
- Electronic structure calculations
- @NSC: VASP, QE, WIEN2k, GPAW, ...

Information / Schedule



https://www.nsc.liu.se/support/Events/VASP_workshop_2024/

<https://enccs.se/events/vasp-best-practices-workshop/>

Wednesday 17th April

10:00 -11:00 Introduction & Basic Theory

11:00 -12:00 VASP - Basics

12:00 -13:00 L u n c h

13:00 -15:00 Hands-on session (guided)

15:00 -17:00 Hands-on session

Thursday 18th April

10:00 -11:00 Running & Performance

11:00 -12:00 Cont., Utilities & Summary

12:00 -13:00 L u n c h

13:00 -15:00 Hands-on session (guided)

15:00 -17:00 Hands-on session

10-15 min breaks every hour

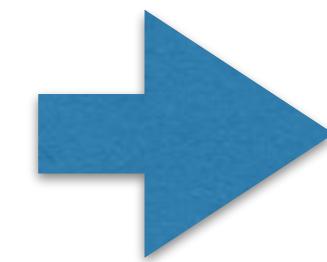
Workshop organization

- Weine Olovsson (NSC) - organizer & presentations
- Yonglei Wang (ENCCS) - organizer & helper
- Diana Iusan (UPPMAX) - helper
- Pavlin Mitev (UPPMAX) - helper
- Wei Li (ENCCS) - helper
- Qiang Li (ENCCS) - helper
- Luis Casillas Trujillo (NSC) - helper

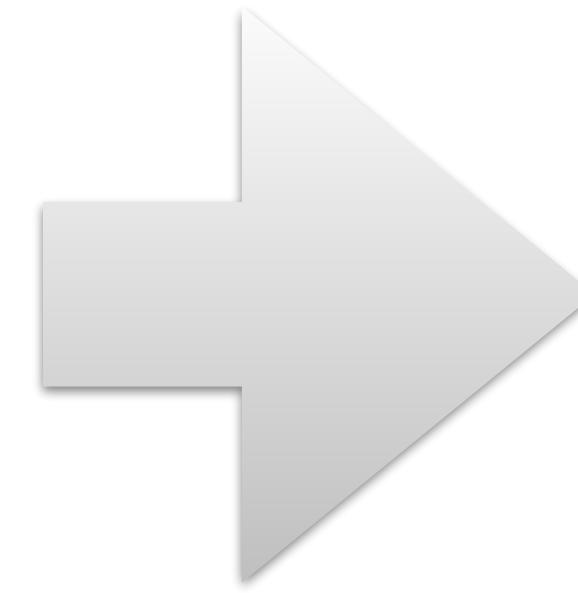
VASP - Best Practices Workshop

- Basic theory (PAW)
- General considerations ...at specific supercomputer centres
- Focus on practical aspects of running VASP
- Influential parameters, NPAR/NCORE, ALGO, NSIM, KPAR, ...
- Benchmarks, examples
- Common problems ... clickable links are underlined

Resources



- Wiki and Manual
Check in detail!
- Examples, tutorials
- Presentations
- Forum



Find all the links:
<https://vasp.at/>

- Also other resources, materials and tools for VASP (see presentation 4.)
- Peter Larsson's old blog at NSC: <https://www.nsc.liu.se/~pla/>
- NSC VASP installations: <https://www.nsc.liu.se/software/installed/tetralith/vasp/>

Questions / trouble @NSC clusters? support@nsc.liu.se

1. Introduction & Basic Theory

Weine Olovsson

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VASP - Best Practices Workshop



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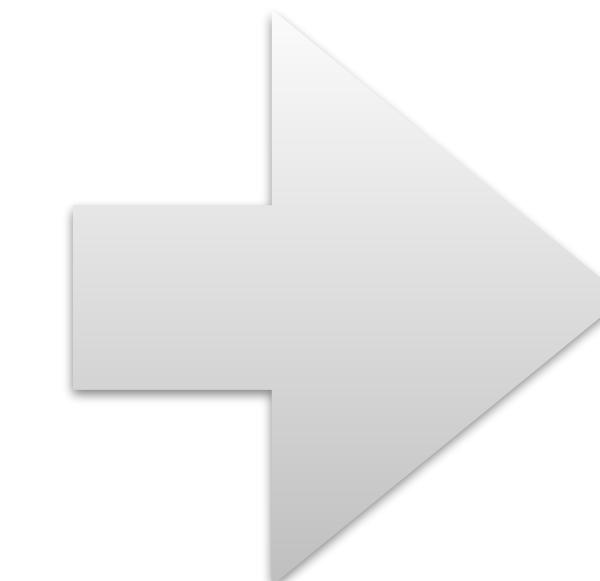
EuroCC National Competence Centre Sweden

<https://enccs.se/>

VASP: short background

- PAW-method
- DFT, **post-DFT** (HSE06, GW, ...)
- Born-Oppenheimer Molecular Dynamics
- **widely used** in Academia/Industry
 - Efforts from **Intel & Nvidia** for optimization
 - 20-25% of Tetralith usage

... clickable links are underlined



<https://vasp.at/>

Schrödinger Equation

Time-independent SE

$$H\Psi = E\Psi,$$

Born-Oppenheimer approx.

$$= 0$$

$$\begin{aligned} H = T + T_n + V_{int} + V_{nn} + V_{ext} &= -\frac{\hbar^2}{2m_e} \sum_i \nabla_i^2 - \boxed{\sum_I \frac{\hbar^2}{2M_I} \nabla_I^2} + \\ &+ \sum_{i \neq j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{1}{2} \sum_{I \neq J} \frac{Z_I Z_J \cdot e^2}{|\mathbf{R}_I - \mathbf{R}_J|} - \sum_{i,J} \frac{Z_J \cdot e^2}{|\mathbf{r}_i - \mathbf{R}_J|}, \end{aligned}$$

solid $\sim 10^{23}$ particles...

How to solve it?

“The general theory of quantum mechanics is now almost complete, ...”

“The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that the exact application of these laws leads to **equations much too complicated to be soluble.**”

“It therefore becomes desirable that **approximate practical methods of applying quantum mechanics should be developed**, which can lead to an explanation of the main features of complex atomic systems without too much computation”

Density Functional Theory (DFT)

Use electron probability density $n(\mathbf{r})$ instead of Ψ ...

(1) The potential V_{ext} of a system is determined uniquely, except for a constant by the ground state density $n(\mathbf{r})$

(2) The total energy functional $E[n]$, for a given V_{ext} , assumes its minimal value for the correct electron density $n(\mathbf{r})$ of the ground state

Ansatz:
$$E_{KS}[n] = \int d^3r V_{\text{ext}}(\mathbf{r}) n(\mathbf{r}) + T_s[n] + E_{xc}[n] + \iint d^3r d^3r' \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|},$$

for *independent* electrons (mean field theory)

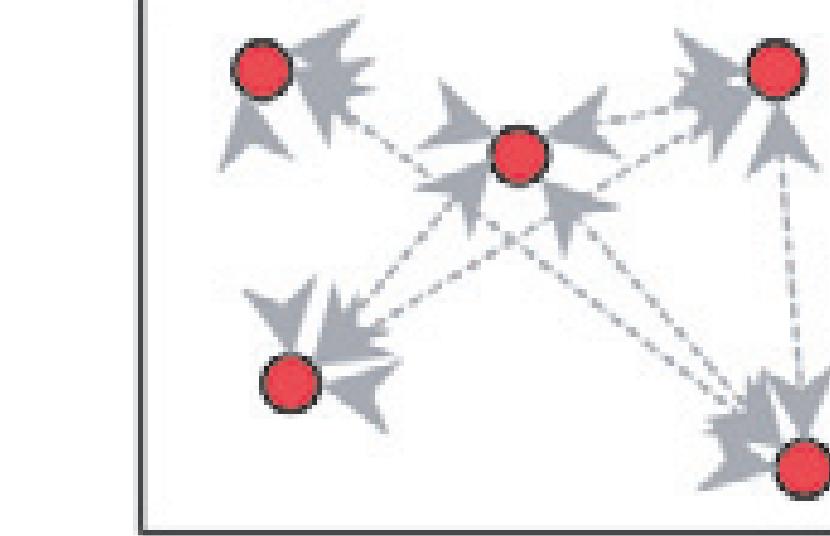
Hohenberg & Kohn, PRL 136, B864 (1964)

Kohn & Sham, PRL 140, A1133 (1965)

Properties of
the system

Hard problem to solve

Schrödinger view

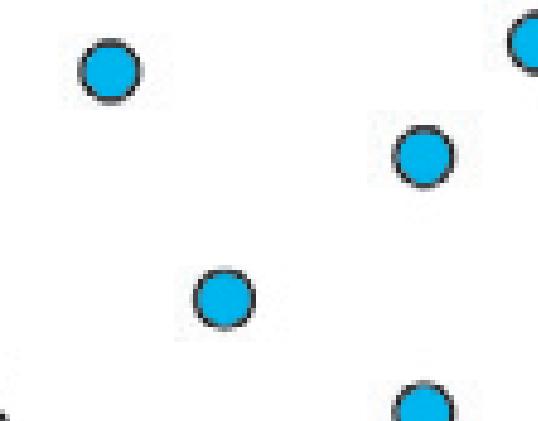


- electron
- ↔ interaction
- external potential

KS

“Easy” problem to solve

DFT view



- Kohn-Sham particle
(non-interacting)
effective potential

Formally
equivalent

$$v_{eff}(\mathbf{r}) = v(\mathbf{r}) + \int \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' + \frac{\delta E_{xc}[n(\mathbf{r})]}{\delta n(\mathbf{r})}$$

LDA, GGA,
Meta-GGA,
Hybrids, ...

Exchange-Correlation Functional

All difficulties now included in the XC-functional, need to find an approximation...

$$E_{xc}^{LDA}[n] = \int d\mathbf{r}^3 n(\mathbf{r}) \epsilon_{xc}(n(\mathbf{r})), \quad \begin{array}{l} \text{Local Density Approximation (LDA)} \\ \text{homogeneous electron gas} \end{array}$$

$$E_{xc}^{GGA}[n] = \int d^3 r n(\mathbf{r}) \epsilon_{xc}(n(\mathbf{r}), |\nabla n|), \quad \text{Generalized Gradient Approximation (GGA)}$$

GGA: PBE, AM05, PBESol, ...

meta-GGA: SCAN, ...

mixing with exact-X: HSE06, ...

many choices, commonly used are e.g. PBE, HSE06, ...

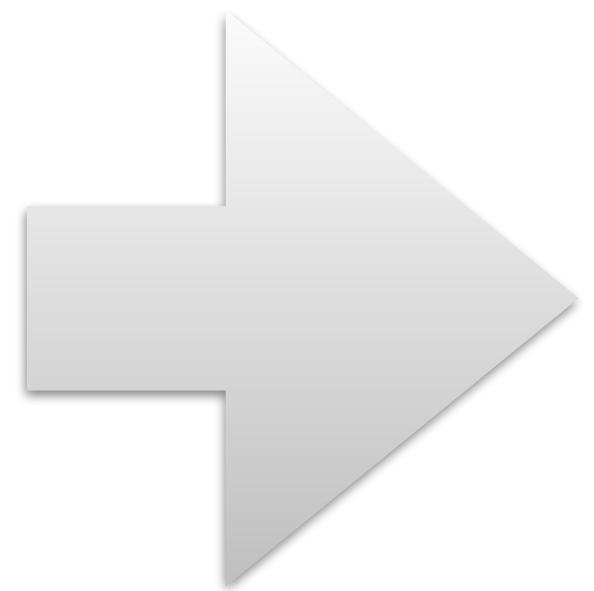
Using periodicity

- The Bloch theorem states that the one-electron wavefunctions obey the equation:

$$\psi_{n\mathbf{k}}(\mathbf{r} + \mathbf{R}) = \psi_{n\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\mathbf{R}}$$

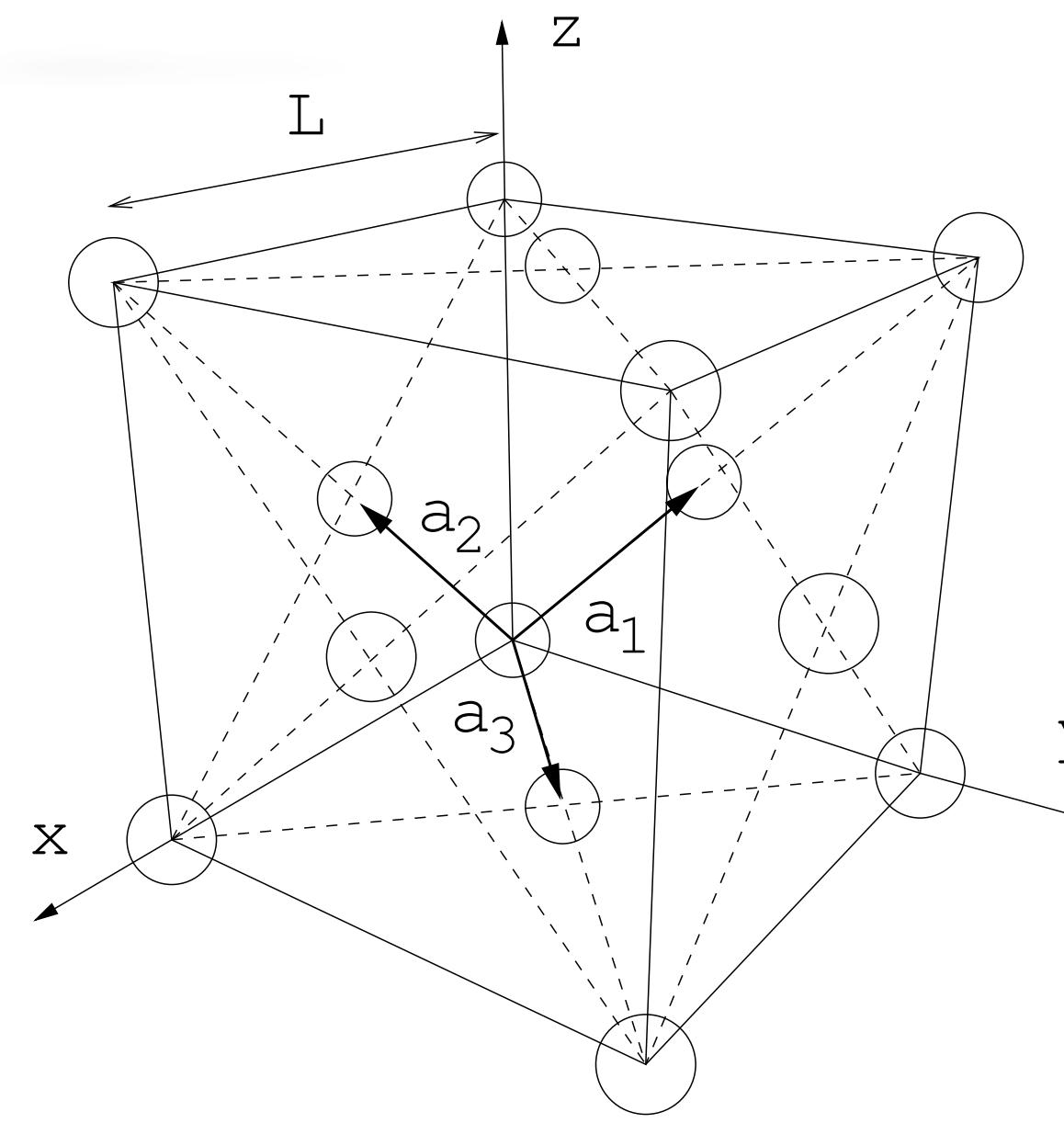
where \mathbf{R} is any translational vector leaving the Hamiltonian invariant.

- \mathbf{k} is usually constrained to lie within the first Brillouin zone in reciprocal space.

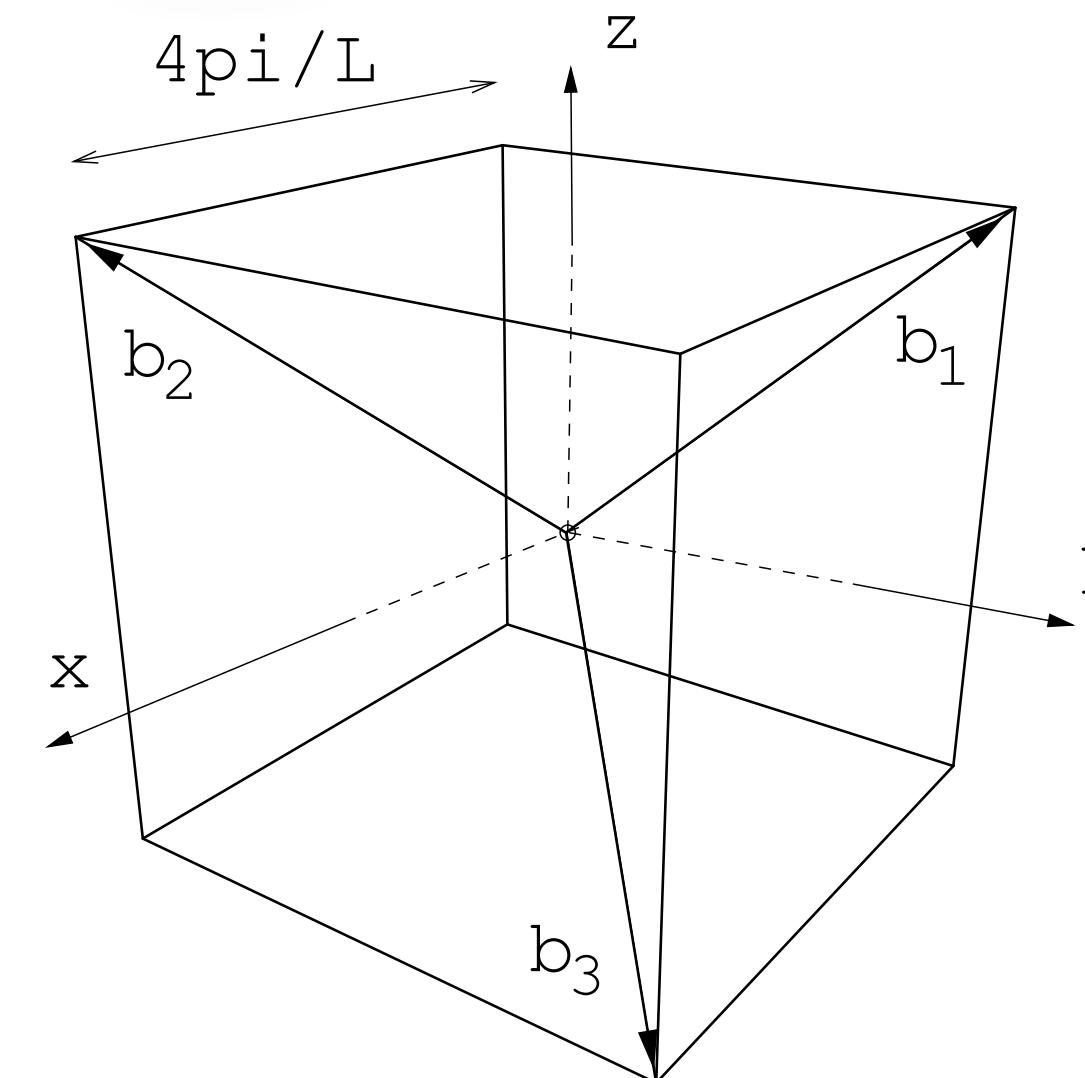


The intractable task of determining $\Psi(\mathbf{r}_1, \dots, \mathbf{r}_N)$ (for $N \sim 10^{23}$) has been reduced to calculating $\psi_{n\mathbf{k}}(\mathbf{r})$ at a discrete set of points $\{\mathbf{k}\}$ in the first BZ, for a number of bands that is of the order of the number of electrons *per unit cell*.

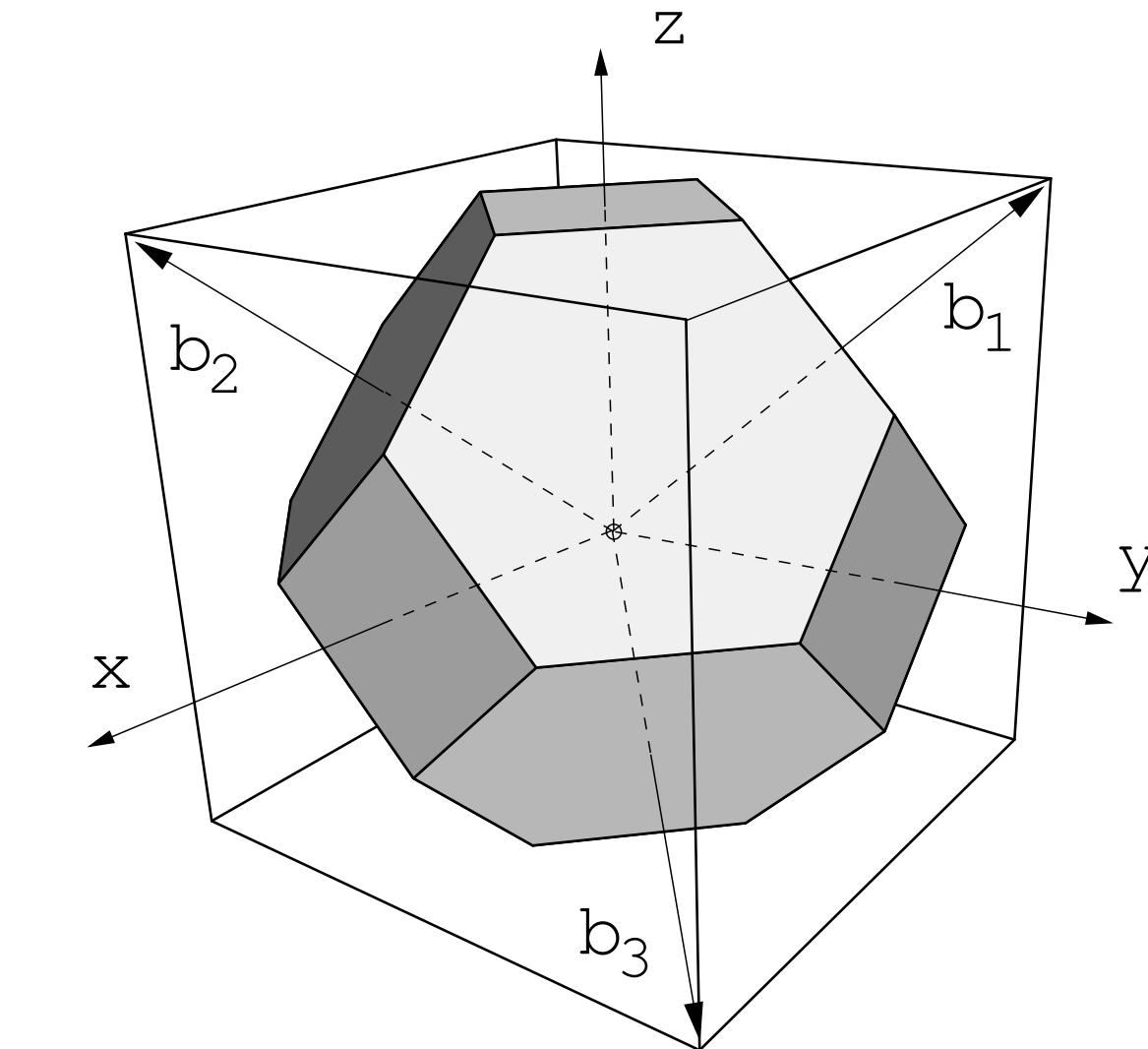
Using periodicity



A



B

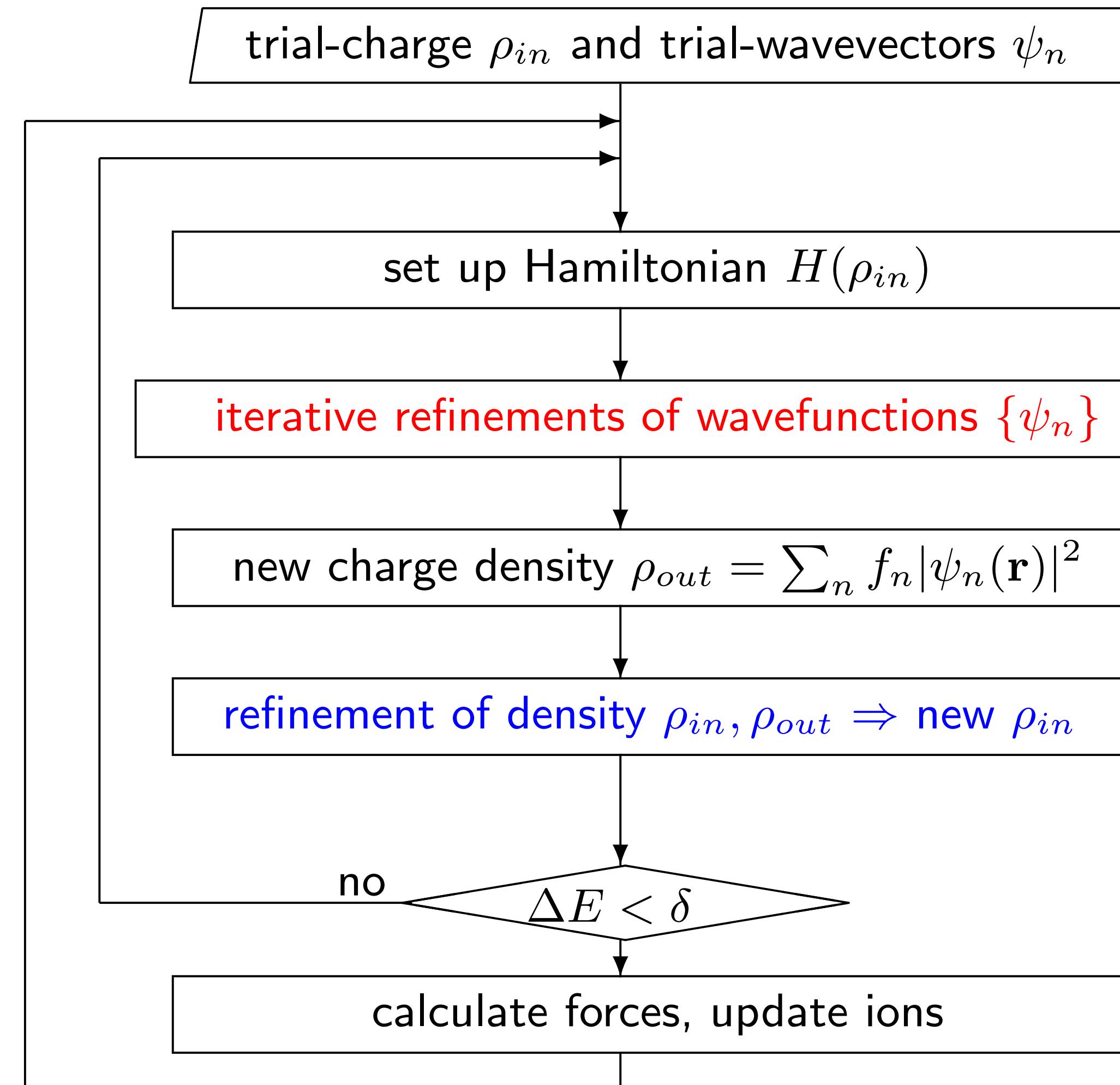


C

$$\mathbf{b}_1 = \frac{2\pi}{\Omega} \mathbf{a}_2 \times \mathbf{a}_3 \quad \mathbf{b}_2 = \frac{2\pi}{\Omega} \mathbf{a}_3 \times \mathbf{a}_1 \quad \mathbf{b}_3 = \frac{2\pi}{\Omega} \mathbf{a}_1 \times \mathbf{a}_2$$

$$\Omega = \mathbf{a}_1 \cdot \mathbf{a}_2 \times \mathbf{a}_3 \quad \mathbf{a}_i \cdot \mathbf{b}_j = 2\pi\delta_{ij}$$

Self-consistent iterations



- two subproblems optimization of $\{\psi_n\}$ and ρ_{in}
- refinement of density: DIIS algorithm
P. Pulay, Chem. Phys. Lett.
73, 393 (1980)
- refinement of wavefunctions: DIIS or Davidson algorithm

Why PAW?

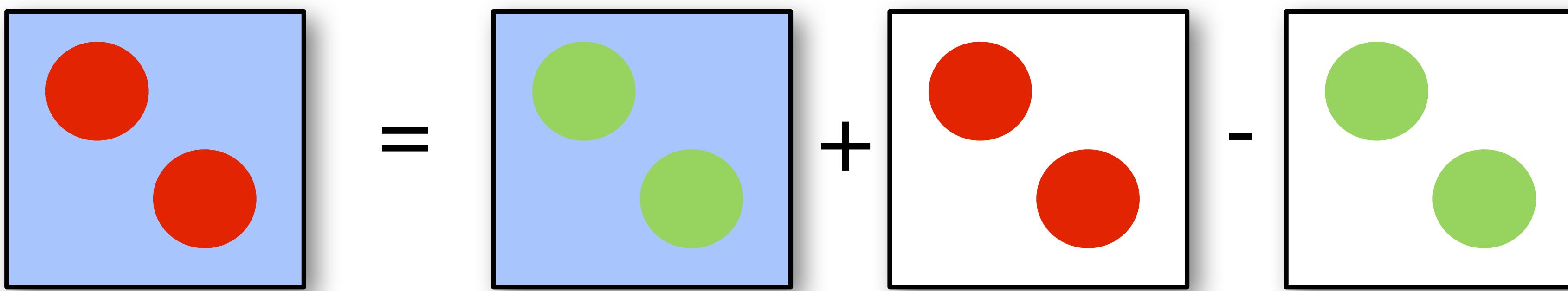
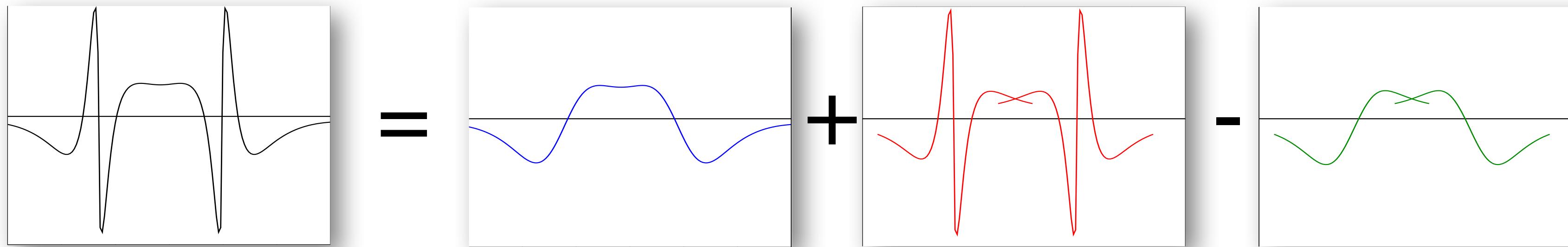
- **Goal:** both **accurate** (LAPW) and **fast** (e.g. USPP) method
- Want to *keep* all-electron (AE) wave function
- Focus on *valence electrons* (frozen core) - chemical bonding
- Fast calculation in *reciprocal space* using FFT (plane waves)
- **Solution:** Projector Augmented Wave (PAW) method

Plane waves & Augmentation

- Rapid wave oscillations close to nucleus
need too many plane waves!
- Strongly localised states at atoms
therefore ->
- Split into *interstitial* and *augmentation* (sphere) regions
smooth pw
- No overlap between spheres (one-centre expansion)
- PAW: Energy and potential *independent* wave functions

PAW Augmentation

$$\underbrace{|\psi\rangle}_{\text{all-electron}} = \underbrace{|\tilde{\psi}\rangle}_{\text{pseudo}} + \underbrace{|\psi^1\rangle}_{\text{1-center, all-el.}} - \underbrace{|\tilde{\psi}^1\rangle}_{\text{1-center, pseudo}}$$
$$\sum_{\alpha} |\phi_{\alpha}\rangle \langle \tilde{p}_{\alpha}| \tilde{\psi} \rangle \quad \sum_{\alpha} |\tilde{\phi}_{\alpha}\rangle \langle \tilde{p}_{\alpha}| \tilde{\psi} \rangle$$



from Blöchl: http://www2.pt.tu-clausthal.de/atp/downloads/lyngby2_paw.pdf

Transformation theory

*True AE wave function and auxiliary PS wf related via **transformation operator**:*

$$|\Psi_n\rangle = \mathcal{T}|\tilde{\Psi}_n\rangle$$

Kohn-Sham equation:

$$H|\Psi_n\rangle = |\Psi_n\rangle\epsilon_n$$

Can write Schrödinger-like equation:

$$\mathcal{T}^\dagger H \mathcal{T} |\tilde{\Psi}_n\rangle = \mathcal{T}^\dagger \mathcal{T} |\tilde{\Psi}_n\rangle\epsilon_n$$

$$\langle A \rangle = \sum_n f_n \langle \Psi_n | A | \Psi_n \rangle = \sum_n f_n \langle \tilde{\Psi}_n | \mathcal{T}^\dagger A \mathcal{T} | \tilde{\Psi}_n \rangle$$

Expectation values can be evaluated for *true AE or auxiliary PS waves*

Transformation operator

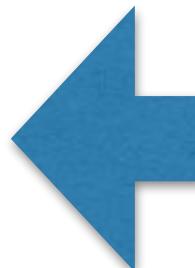
$$\mathcal{T} = 1 + \sum_R \mathcal{S}_R$$

TO unity in *interstitial*, outside
augmentation sphere R

$$\Psi(\mathbf{r}) = \sum_{i \in R} \phi_i(\mathbf{r}) c_i \quad \text{for } |\mathbf{r} - \mathbf{R}_R| < r_{c,R}$$

*Inside sphere R, describe by **AE partial waves**,
undetermined coefficients c_i*

$$\begin{aligned} |\phi_i\rangle &= (1 + S_R)|\tilde{\phi}_i\rangle \quad \text{for } i \in R \\ S_R|\tilde{\phi}_i\rangle &= |\phi_i\rangle - |\tilde{\phi}_i\rangle \end{aligned}$$



Relate AE partial wave with **PS partial wave**,
through *local* transformation operator S

$$\tilde{\Psi}(\mathbf{r}) = \sum_{i \in R} \tilde{\phi}_i(\mathbf{r}) \langle \tilde{p}_i | \tilde{\Psi} \rangle \quad \text{for } |\mathbf{r} - \mathbf{R}_R| < r_{c,R} \quad \text{Expand PS wf in PS partial waves}$$

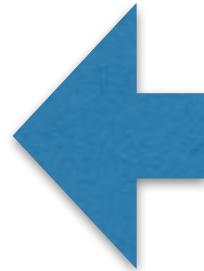
projector function

$$\langle \tilde{p}_i | \tilde{\phi}_j \rangle = \delta_{i,j} \quad \text{for } i, j \in R \quad \text{requirement for above to hold}$$

Transformation operator

$$S_R |\tilde{\Psi}\rangle = \sum_{i \in R} S_R |\tilde{\phi}_i\rangle \langle \tilde{p}_i | \tilde{\Psi}\rangle = \sum_{i \in R} (|\phi_i\rangle - |\tilde{\phi}_i\rangle) \langle \tilde{p}_i | \tilde{\Psi}\rangle \quad \text{From using the previous relations}$$

$$\mathcal{T} = 1 + \sum_i (|\phi_i\rangle - |\tilde{\phi}_i\rangle) \langle \tilde{p}_i |$$



Final expression for the transformation operator

$$|\Psi\rangle = |\tilde{\Psi}\rangle + \sum_i (|\phi_i\rangle - |\tilde{\phi}_i\rangle) \langle \tilde{p}_i | \tilde{\Psi}\rangle = |\tilde{\Psi}_n\rangle + \sum_R (|\Psi_R^1\rangle - |\tilde{\Psi}_R^1\rangle)$$

$$|\Psi_R^1\rangle = \sum_{i \in R} |\phi_i\rangle \langle \tilde{p}_i | \tilde{\Psi}\rangle$$

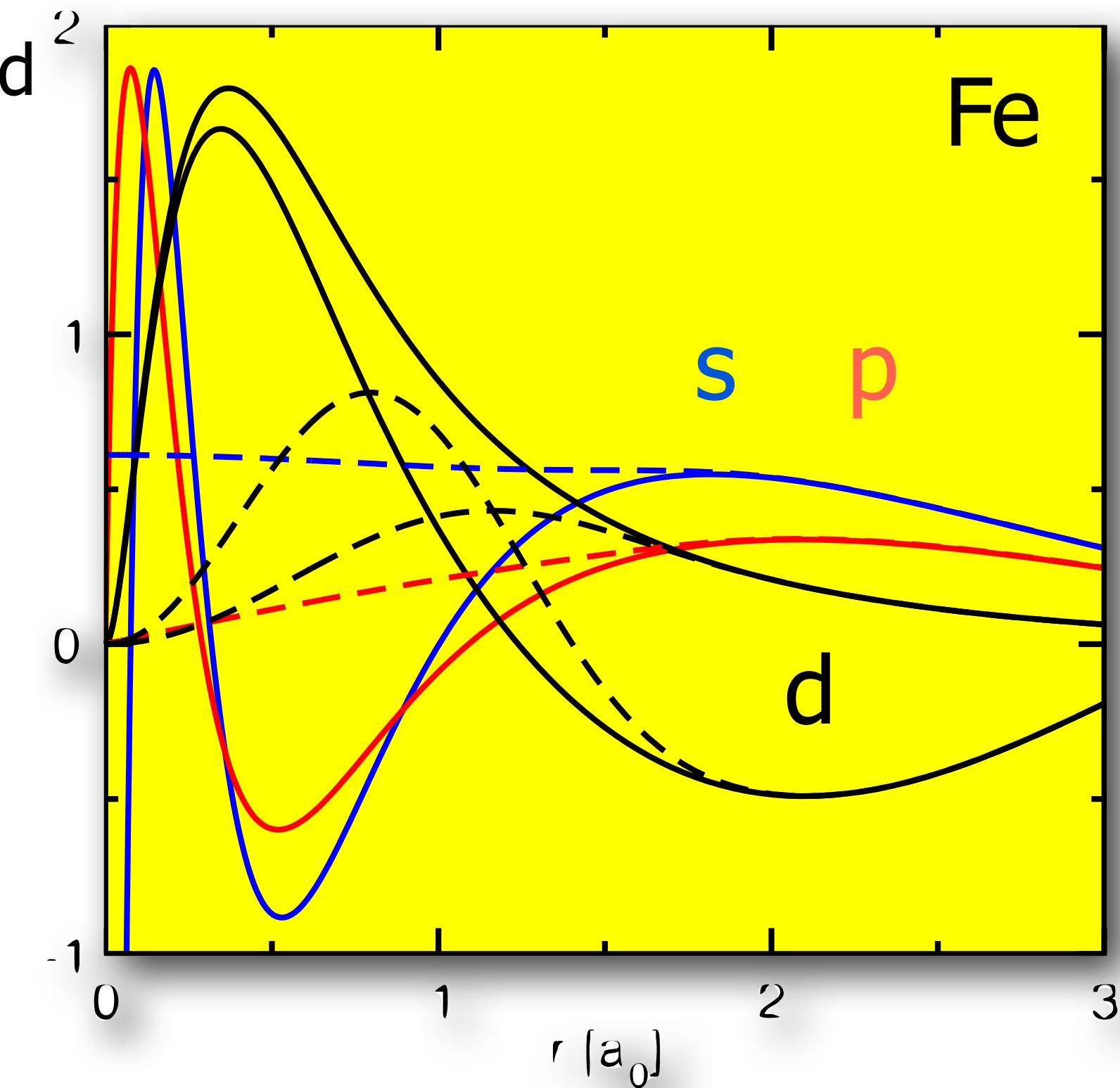
Final expression for the *true AE wf*:

$$|\tilde{\Psi}_R^1\rangle = \sum_{i \in R} |\tilde{\phi}_i\rangle \langle \tilde{p}_i | \tilde{\Psi}\rangle$$

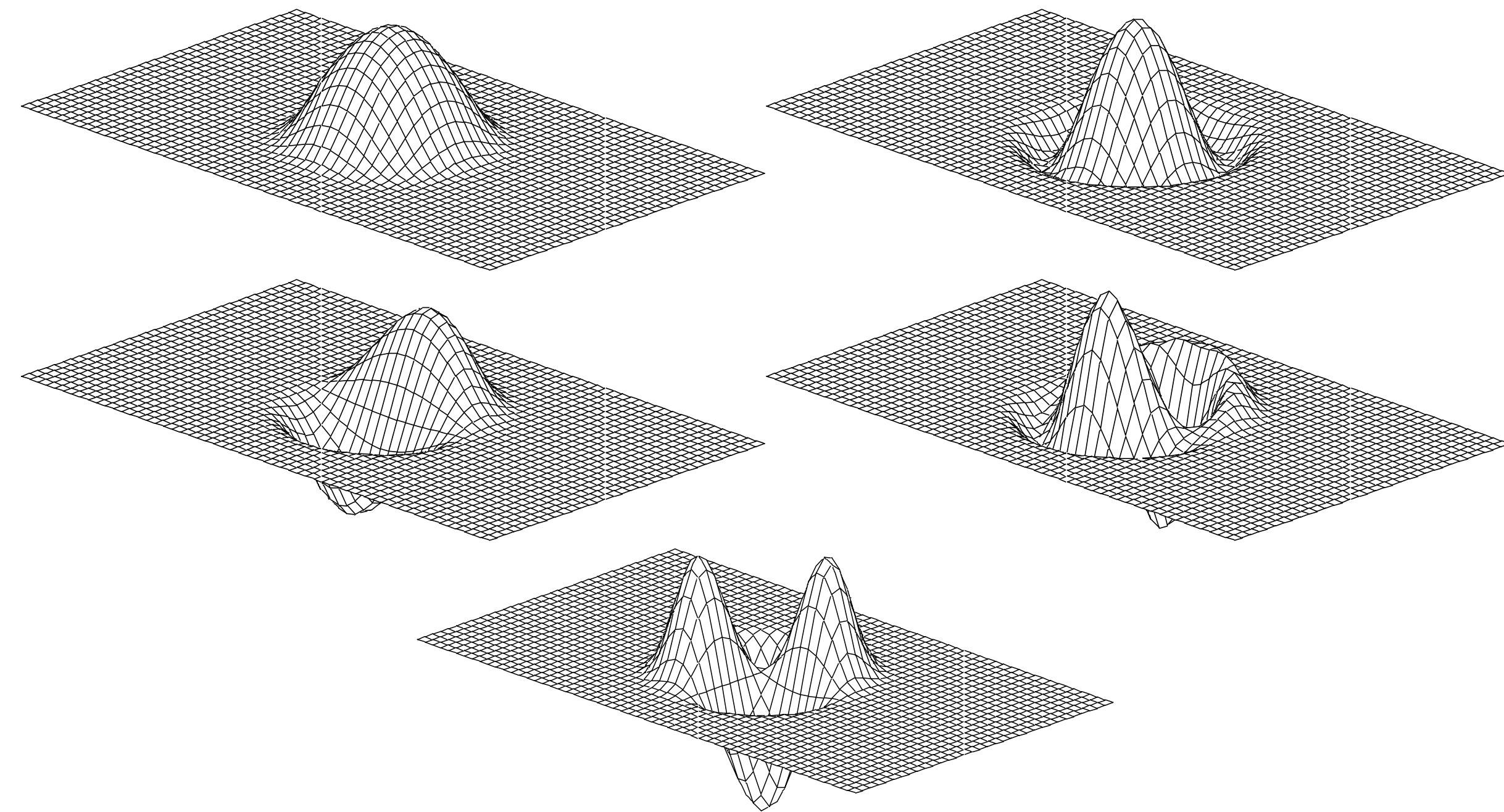
- one PS wf expanded in plane waves
- two atom centred localised functions

Partial waves

- all-electron partial waves $|\phi_\alpha\rangle$
 - integrate Schrödinger equation outward
 - have the correct nodal structure
- pseudo partial waves $|\tilde{\phi}_\alpha\rangle$
 - smooth inside
 - identical to ae partial waves outside
 - $n-n_{\text{core}}$ nodes
 - usually constructed by adjusting an dependent potential



Projector functions



- Localised
- Angular momentum of partial waves

Figure 1: Top: projector functions of the Cl atom for two s-type partial waves, middle: p-type, bottom: d-type.

Wave functions

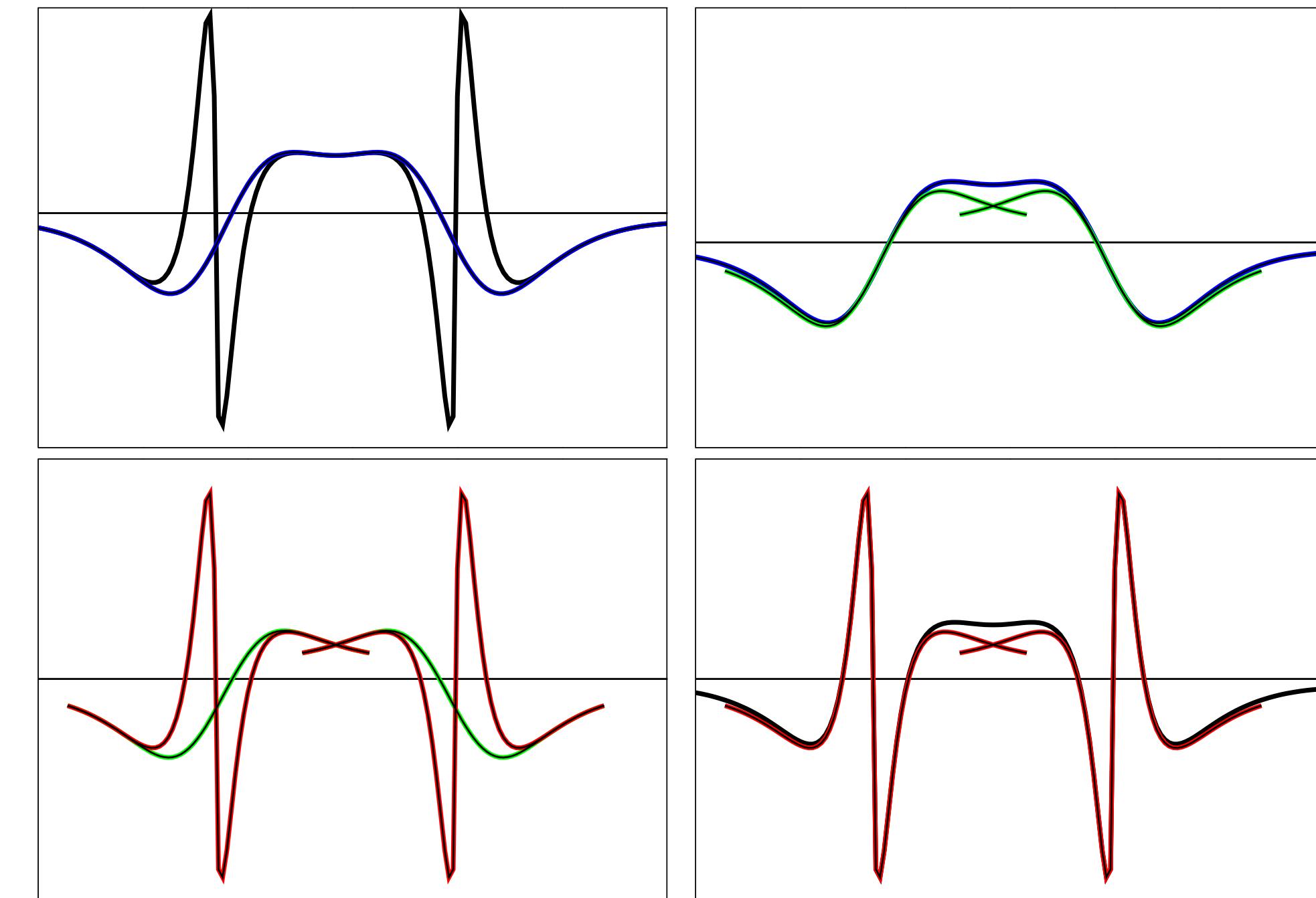
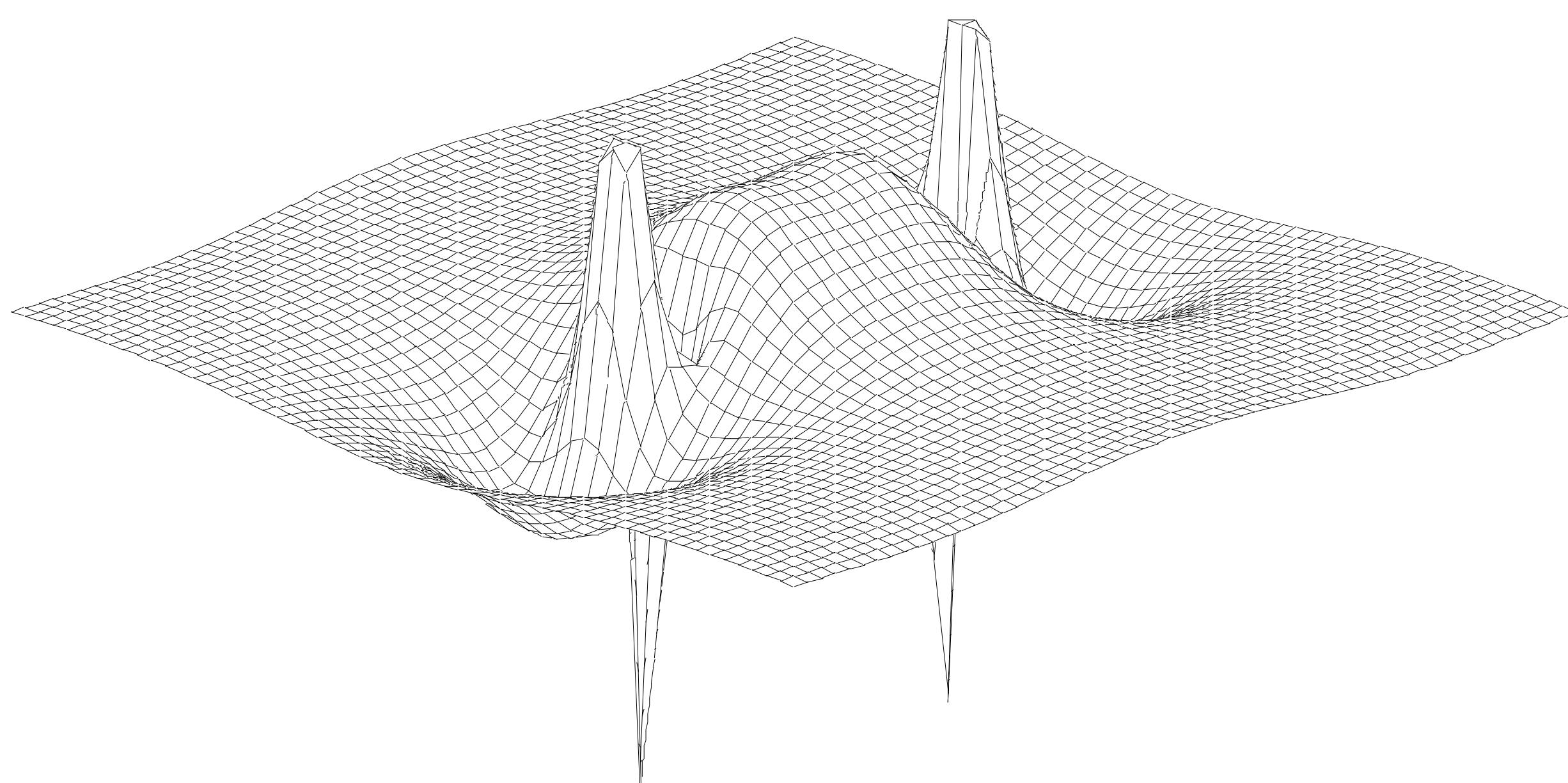


Figure 2: Bonding p- σ orbital of the Cl₂ molecule and its decomposition of the wave function into auxiliary wave function and the two one-center expansions. Top-left: True and auxiliary wave function; top-right: auxiliary wave function and its partial wave expansion; bottom-left: the two partial wave expansions; bottom-right: true wave function and its partial wave expansion.

More examples

AE/PS
proj.

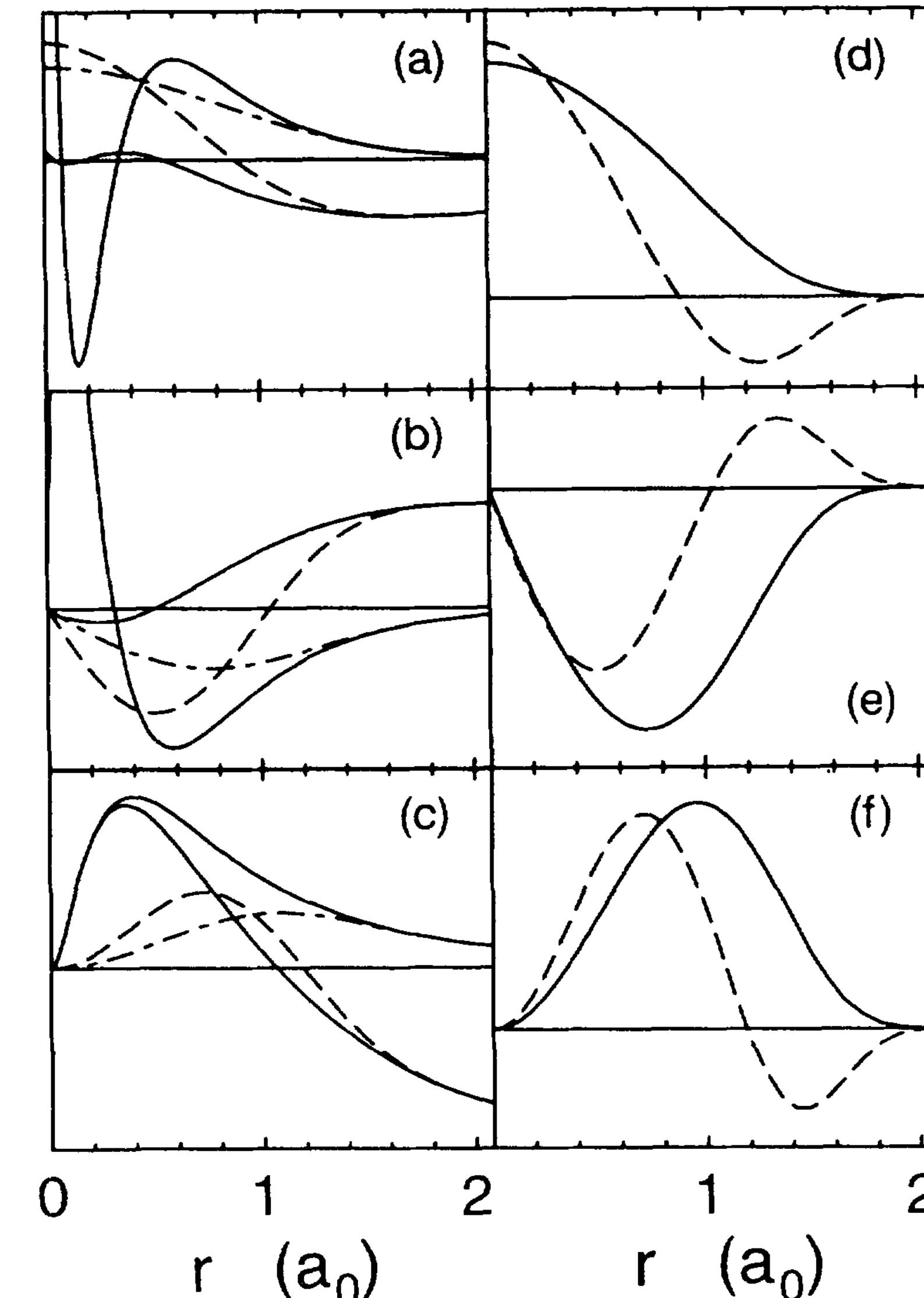


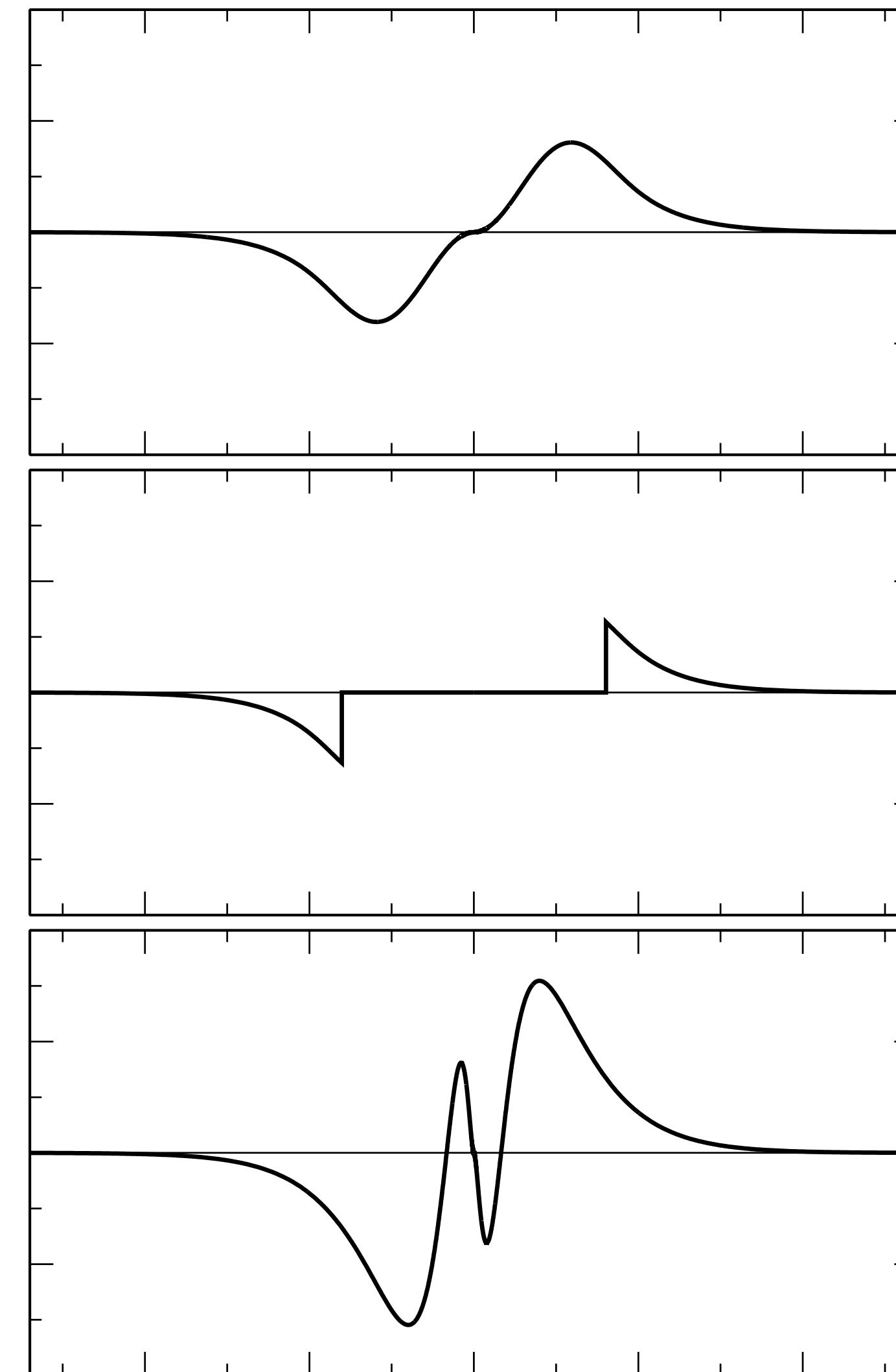
FIG. 1. Partial waves and projectors for Mn. Left panel: AE partial waves (solid lines) and PS partial waves (dashed and dash-dotted lines). The “first” PS partial wave is a dash-dotted line. Right panel: first (solid line) and second (dashed line) projector functions. (a) and (d) show the results for the first and the second partial wave of the s angular momentum channel, respectively, (b) and (e) for the p channel, and (c) and (f) for the d channel. $3s$ and $3p$ functions are treated as valence states. Functions are scaled individually.

PAW Augmentation

$$|\tilde{\psi}_n\rangle$$

$$|\tilde{\psi}_n\rangle - \sum_i |\tilde{\phi}_i\rangle \langle \tilde{p}_i | \tilde{\psi}_n \rangle$$

$$|\tilde{\psi}_n\rangle - \sum_i |\tilde{\phi}_i\rangle \langle \tilde{p}_i | \tilde{\psi}_n \rangle + \sum_i |\phi_i\rangle \langle \tilde{p}_i | \tilde{\psi}_n \rangle$$

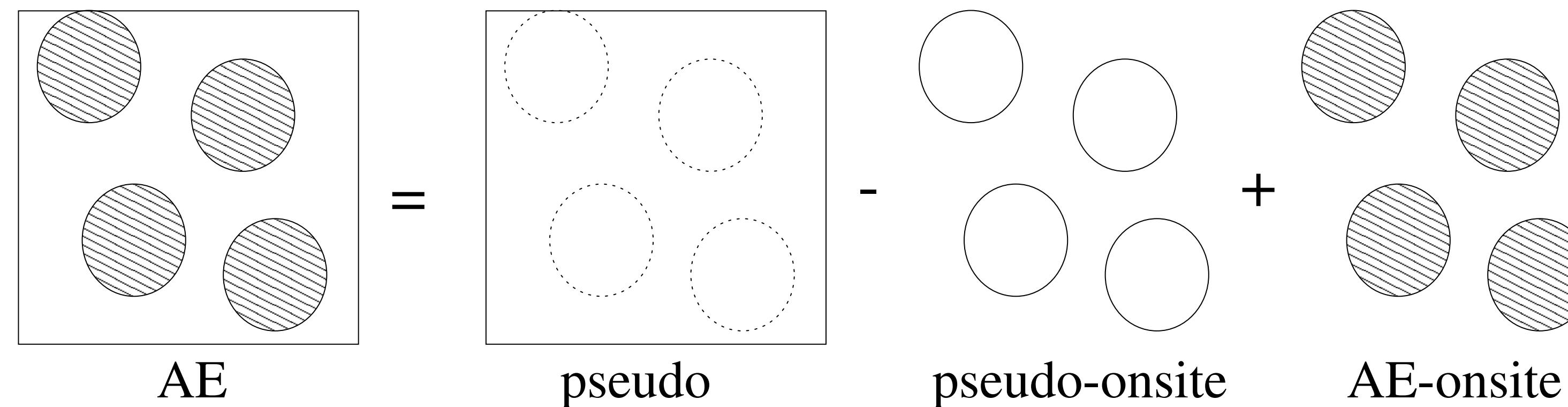


from Marsman: <https://www.vasp.at/mmars/day1.pdf>

PAW Augmentation

- Character of wavefunction: $c_{lme} = \langle \tilde{p}_{lme} | \tilde{\psi}_n \rangle$

$$|\psi_n\rangle = |\tilde{\psi}_n\rangle - \sum |\tilde{\phi}_{lm\epsilon}\rangle c_{lm\epsilon} + \sum |\phi_{lm\epsilon}\rangle c_{lm\epsilon}$$



- Same trick works for
 - Wavefunctions
 - Charge density
 - Kinetic energy
 - Exchange correlation energy
 - Hartree energy

from Marsman: <https://www.vasp.at/mmars/day1.pdf>

Total Energy

$$E = \tilde{E} + E^1 - \tilde{E}^1$$

three terms

PW grid

$$\begin{aligned} \tilde{E} = & \sum_n f_n \langle \tilde{\psi}_n | - \frac{1}{2} \Delta |\tilde{\psi}_n\rangle + E_{xc}[\tilde{\rho} + \hat{\rho} + \tilde{\rho}_c] + \\ & E_H[\tilde{\rho} + \hat{\rho}] + \int v_H[\tilde{\rho}_{Zc}] (\tilde{\rho}(\mathbf{r}) + \hat{\rho}(\mathbf{r})) d^3\mathbf{r} + U(\mathbf{R}, Z_{\text{ion}}) \end{aligned}$$

**local
radial grid**

$$\begin{aligned} \tilde{E}^1 = & \sum_{\text{sites}} \left\{ \sum_{(i,j)} \rho_{ij} \langle \tilde{\phi}_i | - \frac{1}{2} \Delta |\tilde{\phi}_j\rangle + \overline{E_{xc}[\tilde{\rho}^1 + \hat{\rho} + \tilde{\rho}_c]} + \right. \\ & \left. \overline{E_H[\tilde{\rho}^1 + \hat{\rho}]} + \int_{\Omega_r} v_H[\tilde{\rho}_{Zc}] (\tilde{\rho}^1(\mathbf{r}) + \hat{\rho}(\mathbf{r})) d^3\mathbf{r} \right\} \end{aligned}$$

**local
radial grid**

$$\begin{aligned} E^1 = & \sum_{\text{sites}} \left\{ \sum_{(i,j)} \rho_{ij} \langle \phi_i | - \frac{1}{2} \Delta |\phi_j\rangle + \overline{E_{xc}[\rho^1 + \rho_c]} + \right. \\ & \left. \overline{E_H[\rho^1]} + \int_{\Omega_r} v_H[\rho_{Zc}] \rho^1(\mathbf{r}) d^3\mathbf{r} \right\} \end{aligned}$$

What are the approximations?

- Frozen core can be relaxed: Marsman & Kresse, JCP 125, 104101 (2006)
- Plane wave expansion, energy cut-off $\frac{1}{2}|\mathbf{G} + \mathbf{k}|^2 < E_{\text{cutoff}}$
- Partial wave expansion (1-2 per angular momentum)

PAW: Things to note

- *All-electron method* (valence states orthogonal to core)
- Frozen core approximation
- Plane waves: FFT in reciprocal space, **fast calculations**
- **Forces** from total energy expression
- PAW point-of-view: LAPW *special case*, PP an *approximation*

Accuracy

Compare with FPLAPW method (WIEN2k):

| | | | | | | | | | | | | | | | | | | |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| H | | | | | | | | | | | | | | | | | | He |
| 0.1 | | | | | | | | | | | | | | | | | | 0.0 |
| Li | Be | | | | | | | | | | | | | | | | | |
| 0.2 | 0.1 | | | | | | | | | | | | | | | | | |
| Na | Mg | | | | | | | | | | | | | | | | | |
| 0.0 | 0.7 | | | | | | | | | | | | | | | | | |
| K | Ca | Sc | Ti | V | Cr | Mn | Fe | Co | Ni | Cu | Zn | Ga | Ge | As | Se | Br | Kr | |
| 0.1 | 0.2 | 0.4 | 0.9 | 1.3 | 3.1 | 1.4 | 3.4 | 3.4 | 2.0 | 0.4 | 0.3 | 0.2 | 2.4 | 1.7 | 1.5 | 1.5 | 0.1 | |
| Rb | Sr | Y | Zr | Nb | Mo | Tc | Ru | Rh | Pd | Ag | Cd | In | Sn | Sb | Te | I | Xe | |
| 0.1 | 0.1 | 0.5 | 2.7 | 7.3 | 5.5 | 8.3 | 2.3 | 5.4 | 4.4 | 4.1 | 1.4 | 0.4 | 0.2 | 0.1 | 0.5 | 0.9 | 0.1 | |
| Cs | Ba | Lu | Hf | Ta | W | Re | Os | Ir | Pt | Au | Hg | Tl | Pb | Bi | Po | At | Rn | |
| 0.3 | 0.7 | 4.3 | 1.2 | 1.0 | 3.5 | 4.3 | 3.8 | 1.9 | 2.5 | 5.9 | 0.5 | 0.4 | 0.6 | 0.4 | 0.4 | | 0.0 | |

DFT codes using PAW

- VASP license
 - Abinit free
 - Quantum Espresso free
 - GPAW free
 - + more
- Importance of good **potential database**

Refs.

- Good presentations by [Marsman](#) and [Blöchl](#)
- Blöchl PRB **50**, 17953 (1994)
- Blöchl *et al.* <https://arxiv.org/abs/cond-mat/0201015v2>
- Kresse & Joubert PRB **59**, 1758 (1999)
- Holzwarth *et al.* PRB **55**, 2005 (1997)
- Martin, *Electronic Structure*, Chapter 11.1, 13.2

<https://vasp.at/>

