International Summer School on Computational Quantum Materials 2024

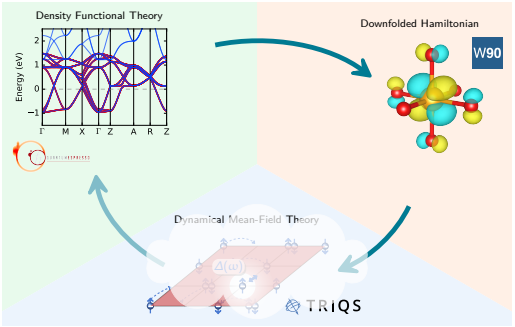
Introduction to Wannier Functions for Materials Simulations

Sophie Beck

22nd May 2024

Localized basis

Goal: transform ab-initio electronic structure into localized basis



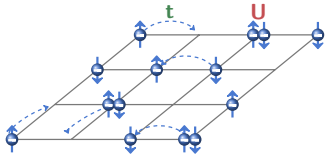
- both conceptual and technical advantages:
  - chemical intuition
  - connection to models
  - local physics, e.g. impurities, defects, local interactions
  - dielectric polarization
  - Wannier interpolation
  - ...
- how to do it in practice?

J. Kuneš, in *Wannier Functions and Construction of Model Hamiltonians* edited by E. Pavarini, E. Koch, D. Vollhardt, A. Liechtenstein (Forschungszentrum, Jülich, 2017)

From many-body to effective one-body problem

electronic Schrödinger equation:

$$\hat{H}\Psi(\mathbf{r}_1, \cdots, \mathbf{r}_N) = \epsilon\Psi(\mathbf{r}_1, ..., \mathbf{r}_N)$$



with

$$\hat{H} = -\sum_i \frac{\hbar^2\nabla_i^2}{2m} + \sum_{i<j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_i^N v_{\text{ext}}(\mathbf{r}_i) = T + U + V_{\text{ext}}$$

in second quantization:

$$\hat{H} = \sum_{ij} t_{ij} c_i^\dagger c_j + \sum_{ijkl} U_{ijkl} c_i^\dagger c_j^\dagger c_l c_k$$

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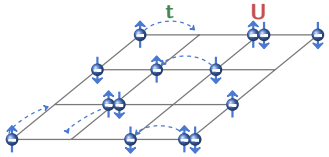

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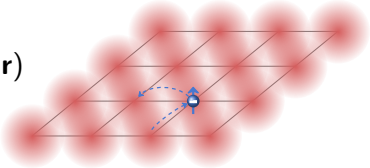
in second quantization:

$$\hat{H} = \sum_{ij} t_{ij} c_i^\dagger c_j + \sum_{ijkl} U_{ijkl} c_i^\dagger c_j^\dagger c_l c_k \rightarrow \hat{H}_{\text{DFT}} = \sum_{ij} \tilde{t}_{ij} c_i^\dagger c_j$$

Effective single-particle picture

Recast full system into a ficticious, auxiliary system of separable Kohn-Sham orbitals $\{\psi_n\}$, that generates the same density as the original one

$$\hat{H}_{\mathrm{KS}}\,\psi_n(\mathbf{r}) = \left[-\frac{\hbar^2}{2m}\nabla^2 + v_{\mathrm{eff}}(\mathbf{r})\right]\psi_n(\mathbf{r}) = \epsilon_n\psi_n(\mathbf{r})$$



$$v_{\mathrm{eff}}(\mathbf{r}) = v_{\mathrm{H}}[\rho](\mathbf{r}) + \frac{\delta E_{\mathrm{XC}}[\rho]}{\delta\rho(\mathbf{r})} + v_{\mathrm{ext}}(\mathbf{r})$$

- solution is found self-consistently
- exchange-correlation potential is the only unknown
- Kohn-Sham orbitals have little physical meaning

$$\rightarrow \hat{H}_{\mathrm{KS}} = \sum_{ij} \tilde{t}_{ij} c_i^\dagger c_j$$

The Bloch theorem

For electrons in a periodic potential the solutions to the Schrödinger equation are Bloch functions:

$$\hat{H}_{\mathrm{KS}}\,\psi_{n\mathbf{k}}(\mathbf{r}) = \epsilon_{n\mathbf{k}}\psi_{n\mathbf{k}}(\mathbf{r})$$

with

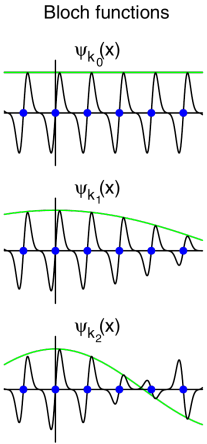
$$\psi_{n\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}u_{n\mathbf{k}}(\mathbf{r}) \qquad \text{with} \qquad u_{n\mathbf{k}}(\mathbf{r}) = u_{n\mathbf{k}}(\mathbf{r} + \hat{T}_i)$$

A few remarks:

- $[\hat{H}, \hat{T}_i] = 0$, block diagonalization
- discrete spectrum $n \in \mathbb{Z}$
- $\mathbf{k} \in \mathrm{BZ}$, continuous variable for the infinite crystal
- $\psi_{n\mathbf{k}}(\mathbf{r})$ orthonormal
- gauge freedom $\psi_{n\mathbf{k}}(\mathbf{r}) \to e^{i\phi(\mathbf{k})}\psi_{n\mathbf{k}}(\mathbf{r})$
- $\psi_{n\mathbf{k}}(\mathbf{r})$ can be smooth function of $\mathbf{k}$

F. Bloch, *Über die Quantenmechanik der Elektronen in Kristallgittern*, Zeitschrift für Physik 52, 555 (1929)Â ·

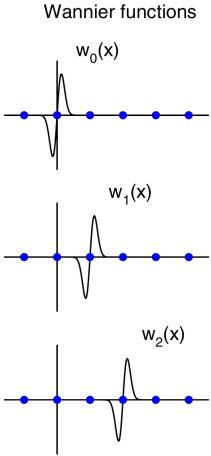
From Bloch states to Wannier functions



- Bloch functions at different **k** have different envelope functions $e^{i\mathbf{k}\cdot\mathbf{r}}$

$$\psi_{n\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r})$$

- build localized basis by superposing Bloch functions
- requirements:
  - should span same subspace
  - should be orthonormal basis



N. Marzari *et al.*, Rev. Mod. Phys. 84, 1419-1475 (2012)

From Bloch states to Wannier functions

- for one band n:

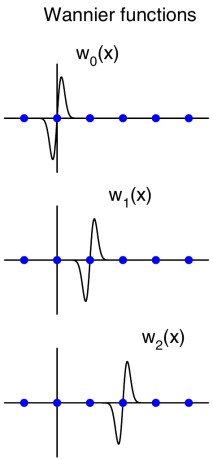
$$w_{n\mathbf{0}}(\mathbf{r}) = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} d\mathbf{k}\, \psi_{n\mathbf{k}}(\mathbf{r})$$

- for multiple Wannier functions:

$$w_{n\mathbf{R}}(\mathbf{r}) = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} d\mathbf{k}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}\psi_{n\mathbf{k}}(\mathbf{r})$$

with $w_{n\mathbf{R}}(\mathbf{r}) = \langle \mathbf{r}|\mathbf{R}n\rangle = w_{n\mathbf{0}}(\mathbf{r}-\mathbf{R})$, or

$$|\mathbf{R}n\rangle = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} d\mathbf{k}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}|\psi_{n\mathbf{k}}\rangle$$



G. H. Wannier, Phys. Rev. 52, 191-197 (1937)

Properties of Wannier functions

Properties of $|\mathbf{R}n\rangle$:

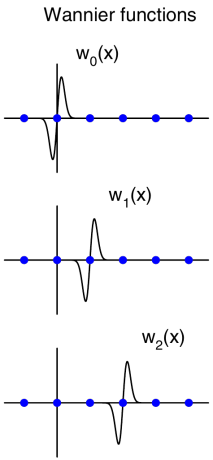
- they form an orthonormal set:

$$\langle \mathbf{R}n|\mathbf{R}'m\rangle = \delta_{\mathbf{R}\mathbf{R}'}\delta_{nm}$$

- $\{|\mathbf{R}n\rangle\}$ span the same space as $\{|\psi_{n\mathbf{k}}\rangle\}$:

$$P = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} \mathrm{d}\mathbf{k}\, |\psi_{n\mathbf{k}}\rangle\langle\psi_{n\mathbf{k}}| = \sum_{\mathbf{R}} |\mathbf{R}n\rangle\langle\mathbf{R}n|$$

- they are translational images of $|\mathbf{0}n\rangle$



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Localization of Wannier functions

Properties of $|\mathbf{R}n\rangle$:

- localized *to some extent* in real space

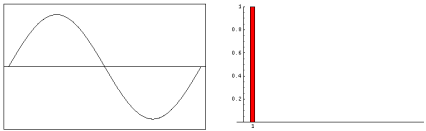
Fourier transform

$$|\mathbf{R}n\rangle = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} d\mathbf{k}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}|\psi_{n\mathbf{k}}\rangle$$

Fourier series (inverse Fourier transform)

$$|\psi_{n\mathbf{k}}\rangle = \sum_{\mathbf{R}} \mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}}\,|\mathbf{R}n\rangle$$

- smooth, periodic function has Fourier coefficients that decay rapidly
- "gauge freedom": $|\psi^{\mathrm{W}}_{n\mathbf{k}}\rangle = e^{i\varphi_n(\mathbf{k})}|\psi_{n\mathbf{k}}\rangle$
- choose $\varphi_n(\mathbf{k})$ so that $|\psi^{\mathrm{W}}_{n\mathbf{k}}\rangle$ are smooth in $\mathbf{k}$



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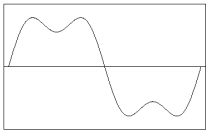

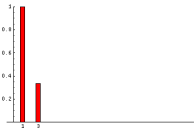

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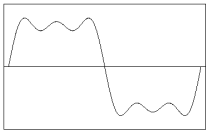

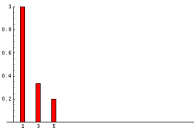

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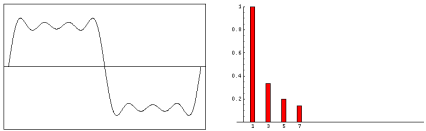

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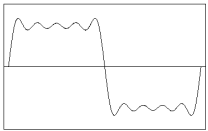

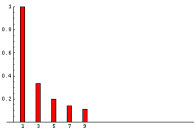

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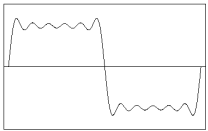

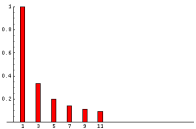

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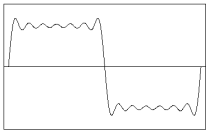

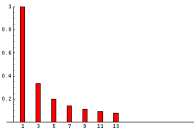

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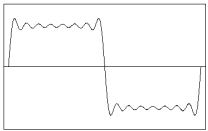

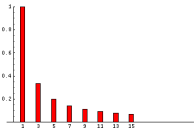

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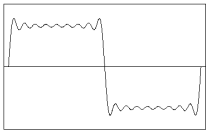

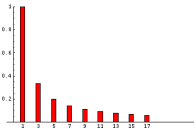

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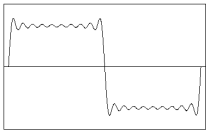
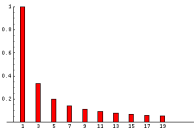




https://mriquestions.com/fourier-transform-ft.html

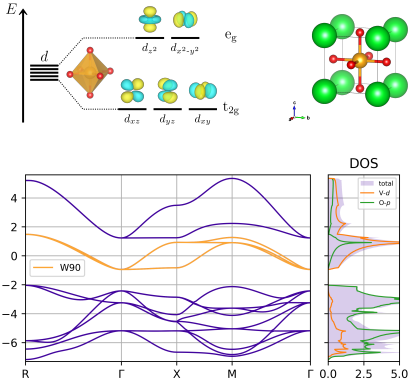
Wannier functions - multiple bands

- "gauge freedom" for multiple bands:

$$|\mathbf{R}j\rangle = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} d\mathbf{k}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{R}}|\psi_{j\mathbf{k}}\rangle$$

- no one-to-one correspondence band $n \leftrightarrow$ orb. j
- Wannier functions strongly nonunique!
- $U_{\mathbf{k}}$ encode the *gauge selection*
- how to choose the unitary rotations to get localized WFs?

Example: $SrVO_3$



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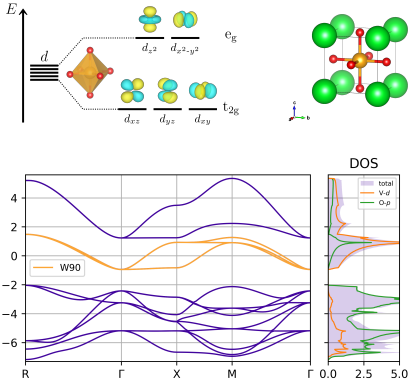

How to get Wannier functions in practice

Projection method:

- start with localized trial orbitals $g_n(\mathbf{r})$:

$$|\phi_{n\mathbf{k}}\rangle = \sum_{m=1}^{J} A_{\mathbf{k},mn} |\psi_{m\mathbf{k}}\rangle \quad \text{with} \quad A_{\mathbf{k},mn} = \langle \psi_{m\mathbf{k}} | g_n \rangle \quad \texttt{(Wannier90)}$$

- construct Löwdin-orthonormalized Bloch-like states:

$$|\tilde{\psi}_{n\mathbf{k}}\rangle = \sum_{m=1}^{J} |\phi_{n\mathbf{k}}\rangle \, S^{-1/2}_{\mathbf{k},mn} \quad \text{with} \quad S_{\mathbf{k},mn} = (A^\dagger A)_{\mathbf{k},mn}$$

- the $|\tilde{\psi}_{n\mathbf{k}}\rangle$ are smooth in $\mathbf{k}$, leading to well-localized Wannier functions
- advantages: simple, they retain the symmetry of the trial orbitals
- disadvantages: cases without good guesses, e.g. molecular orbitals, low-symmetry

N. Marzari *et al.*, Rev. Mod. Phys. 84, 1419-1475 (2012)

Maximally localized Wannier functions (MV method)

- Marzari and Vanderbilt scheme: choose optimal $U_{\mathbf{k}}$ that minimize spread functional

$$\Omega = \sum_{j=1}^{J} \left[\langle \mathbf{0}j \,|\, r^2 \,|\, \mathbf{0}j \rangle \; - \; \left| \langle \mathbf{0}j \,|\, \mathbf{r} \,|\, \mathbf{0}j \rangle \right|^2 \right]$$

- iterative minimzation using steepest-descents or conjugate-gradient method
- Blount identities provide matrix elements of position operator in Wannier basis:

$$\langle \mathbf{R}i | \mathbf{r} | \mathbf{0}j \rangle = i \, \frac{V}{(2\pi)^3} \int d\mathbf{k} \, e^{i\mathbf{k}\cdot\mathbf{R}} \langle u_{i\mathbf{k}} | \nabla_{\mathbf{k}} | u_{j\mathbf{k}} \rangle$$

- can be recast in terms of overlap matrices:

$$M_{ij}^{(\mathbf{k},\mathbf{b})} = \langle u_{i\mathbf{k}} | u_{j\mathbf{k}+\mathbf{b}} \rangle \qquad \texttt{(Wannier90)}$$

N. Marzari and D. Vanderbilt, Phys. Rev. B 56, 12847 (1997)

E.I. Blount, "Formalisms of band theory," in Solid State Phys., Vol. 13 (Elsevier) p. 305. (1962)

Disentanglement procedure (SMV scheme)

- previous concepts extend to non-isolated ("entangled") groups of bands
- identify J-dim. Bloch manifold from a larger set of $\mathcal{J}_{\mathbf{k}}$ Bloch eigenstates (*subspace selection*):

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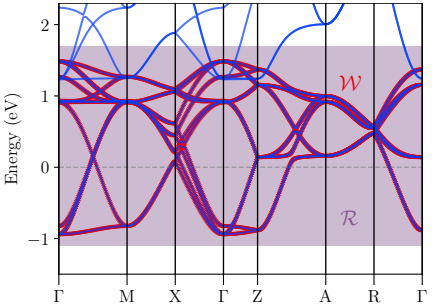

I. Souza, N. Marzari, and D. Vanderbilt, Phys. Rev. B 65, 035109 (2001)

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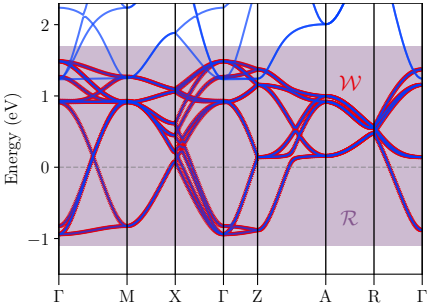

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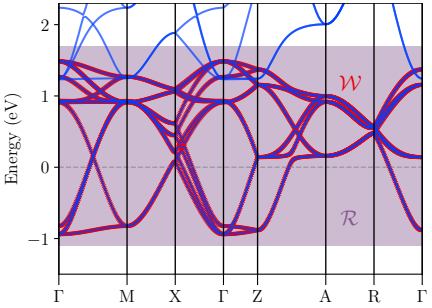

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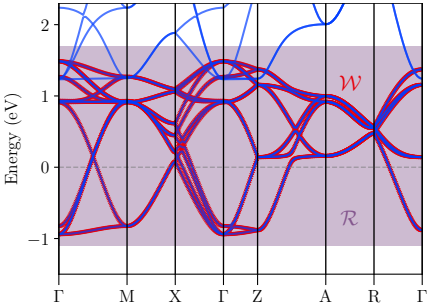

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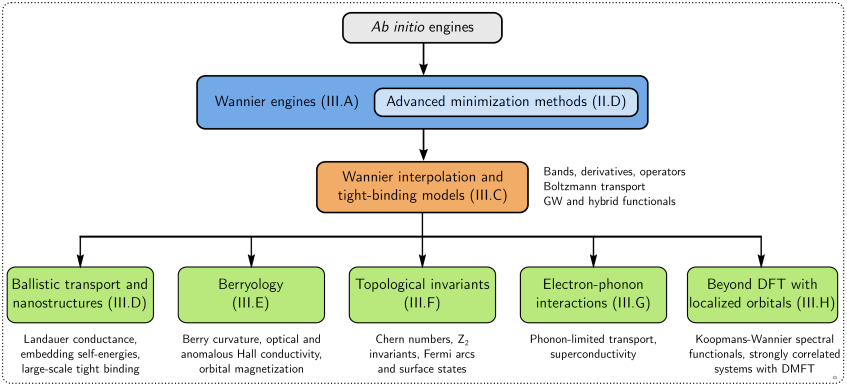
Major applications of Wannier functions

- Interpolation
- Geometry and Topology
- Advanced electronic-structure methods



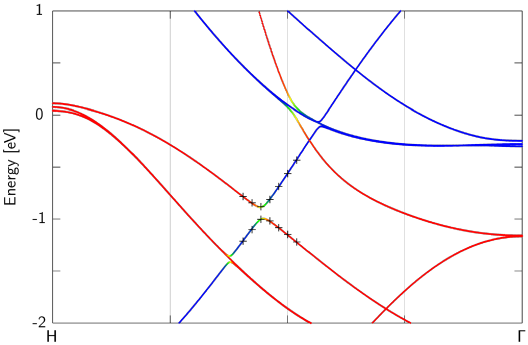
A. Marrazzo, SB *et al.*, arxiv:2312.10769 (2023)

Wannier interpolation and tight-binding models

- $\{|\mathbf{R}n\rangle\}$ span the same space as $\{|\psi_{n\mathbf{k}}\rangle\}$
- efficient interpolation in reciprocal space important for BZ integrals

$$\bar{f}_n = \frac{1}{N_k} \sum_{\mathbf{k}} f_n(\mathbf{k}) \to \frac{1}{\Omega} \int_{\mathrm{BZ}} \mathrm{d}\mathbf{k}\, f_i(\mathbf{k})$$

- reproduce correct band connectivity



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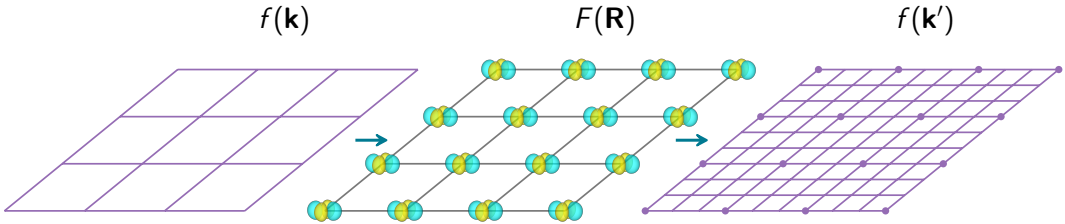

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$$= \left[V_{\mathbf{k}}^{\dagger}(\mathbf{k}) H(\mathbf{k}) V_{\mathbf{k}} \right]_{nm}$$

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$$H_{nm}(\mathbf{k}) = \epsilon_{n\mathbf{k}} \delta_{nm}$$

J. R. Yates, X. Wang, D. Vanderbilt, and I. Souza, Phys. Rev. B 75, 195121 (2007)

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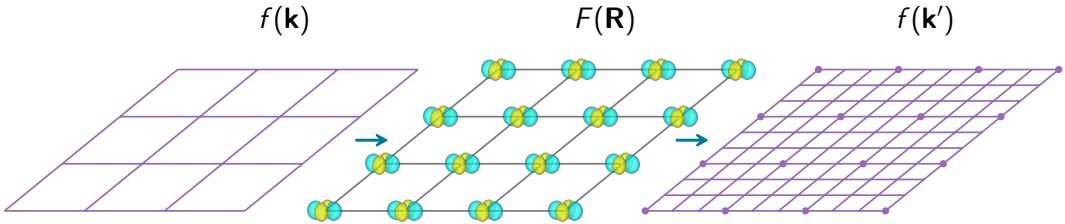

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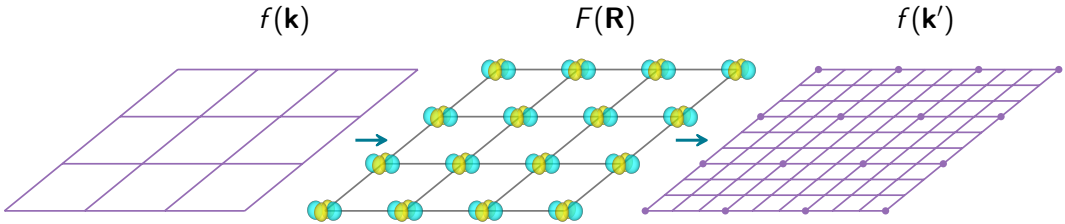

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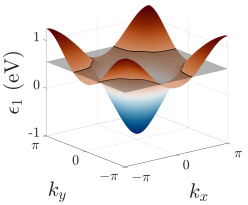

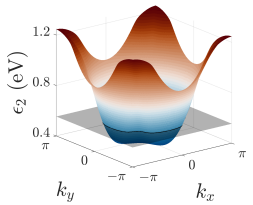

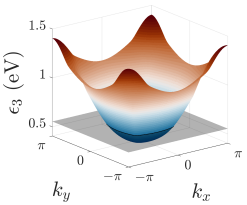

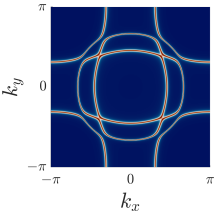

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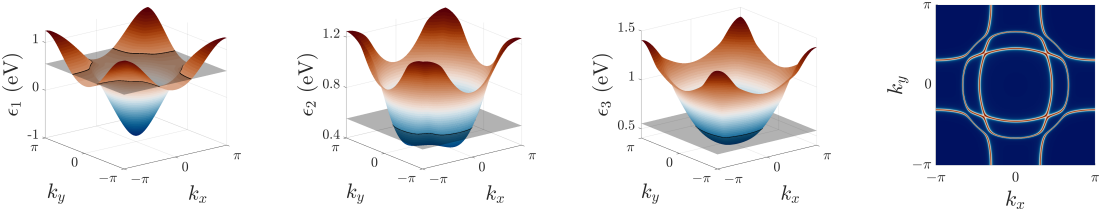

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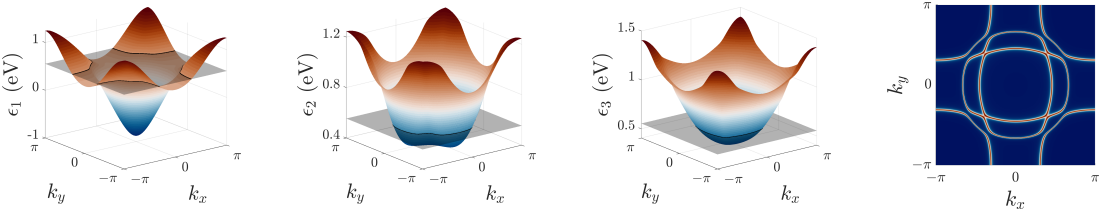

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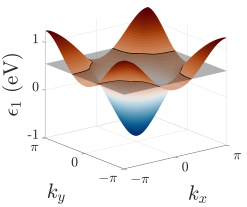

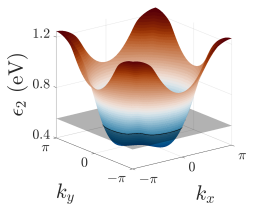

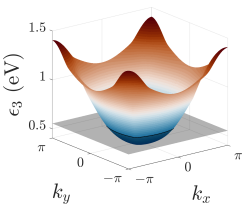

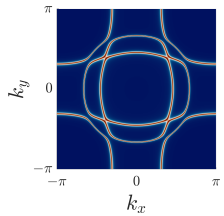

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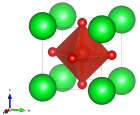
Adaptive Brillouin zone integration - DOS of $SrVO_3$

- density of states (DOS):



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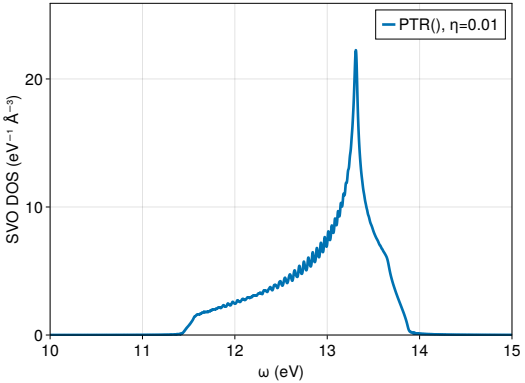
- comparison of default (PTR) versus new (IAI) algorithm
- user-provided error tolerance eliminates convergence tests
- available in `AutoBZ.jl` package



L. Van Muñoz, SB, and J.Kaye, *in preparation* (2024)

https://github.com/lxvm/AutoBZCore.jl/tree/main/aps_example

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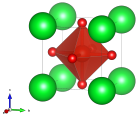

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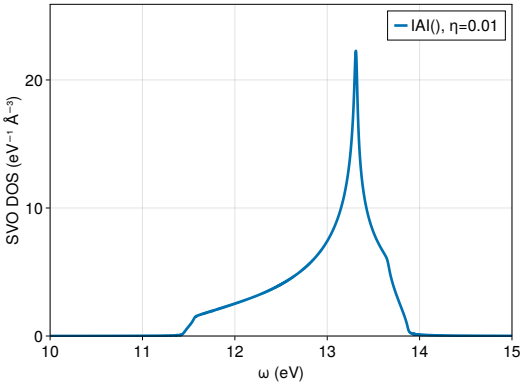

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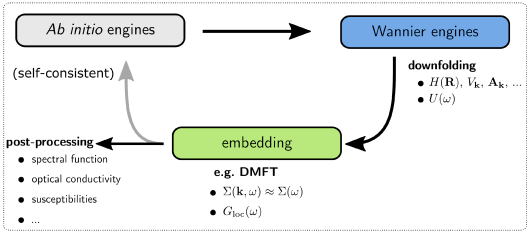
Beyond DFT with localized orbitals

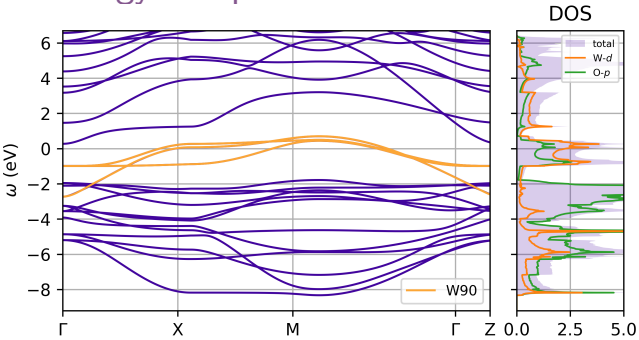


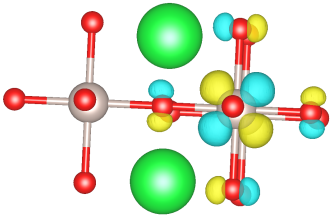
- **Idea**: DFT description good for most states, many-body (MB) treatment for low-energy states
- **Approach**: partition the total Hilbert space efficiently using WFs
- charge self-consistency important (for some systems)

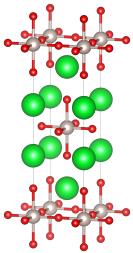
→ see lecture by O. Parcollet

A. Marrazzo, SB *et al.*, arxiv:2312.10769 (2023)

Low-energy subspace with Wannier functions



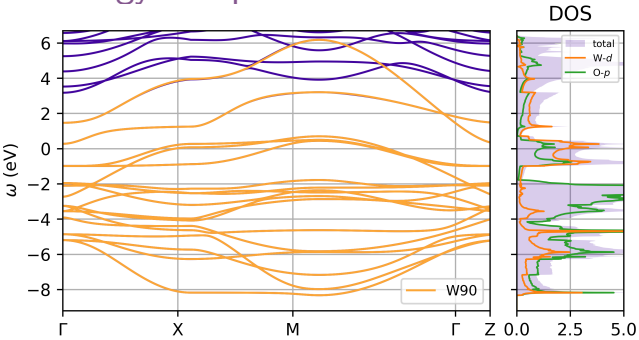


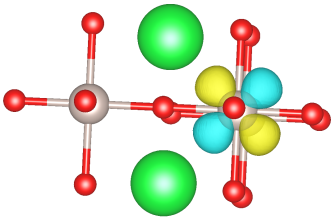


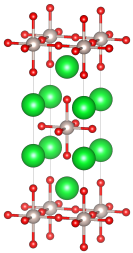
Degree of localization...

- depends on energy window and contained states (hybridization)
- affects difficulty of MB calculations, value/form of local interaction
- Example: 3 t_{2g} orbitals versus 5+9 *dp* orbitals in Sr_2RuO_4

Low-energy subspace with Wannier functions







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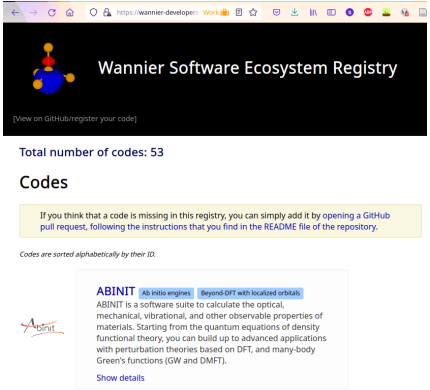
Wannier functions ecosystem

Ecosystem based on:

- theoretical advancement
  - e.g. automation workflows, novel localization algorithm, ...
- development of open-source software packages
  - Wannier Software Ecosystem Registry*
- user support†
  - documentation, tutorials, mailing list, schools, developers meeting, ...



*https://wannier-developers.github.io/wannier-ecosystem-registry/

†https://wannier.org/support

Wannier functions for the practitioner using `Wannier90`

Building Wannier functions:

- ingredients from an electronic structure calculation:
  - overlap between Bloch states (`wannier90.mmn`): $M_{ij}^{(\mathbf{k},\mathbf{b})} = \langle u_{i\mathbf{k}} | u_{j\mathbf{k}+\mathbf{b}} \rangle$
  - projection onto trial localized orbitals (`wannier90.amn`): $A_{mn}^{(\mathbf{k})} = \langle \psi_{m\mathbf{k}} | g_n \rangle$
- user-defined input (`wannier90.win`):
  - trial orbitals, disentanglement parameters, ...

Other quantities:

- hoppings (input: `wannier90.eig` $\rightarrow$ output: `wannier90_hr.dat`)
- additional ingredients based on quantity of interest...

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