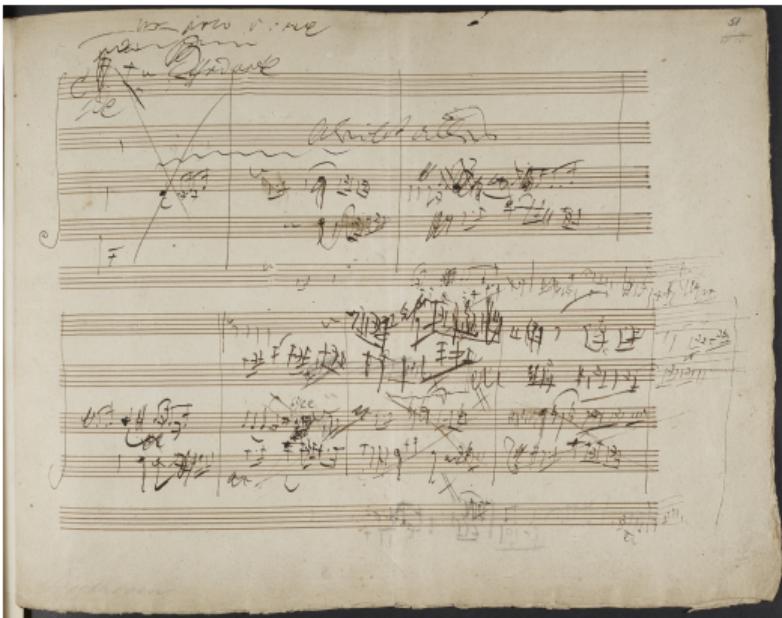


Beethoven's late string quartets

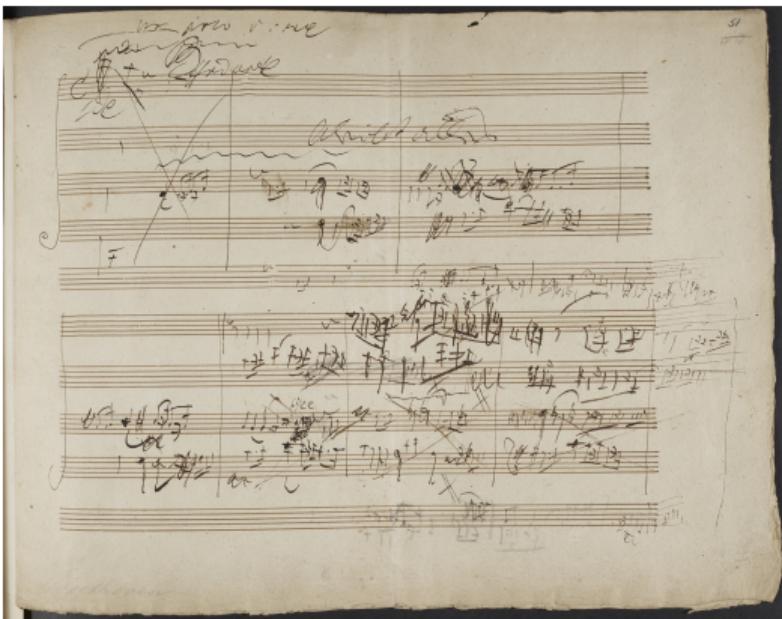
String Quartet No. 14 - Adagio ma non troppo e molto espressivo



[click here for video](#)

Beethoven's late string quartets

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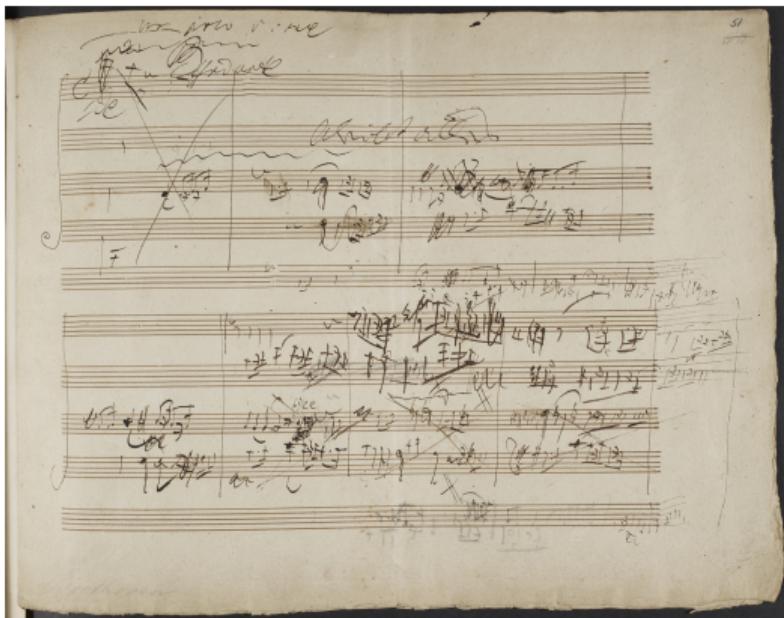


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“indecipherable, uncorrected horrors” – Spohr

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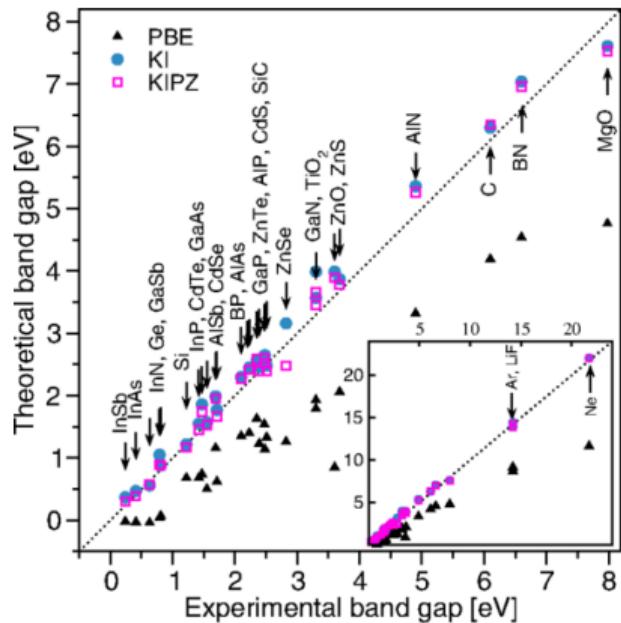
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“indecipherable, uncorrected horrors” – Spohr
“After this, what is left for us to write?” – Schubert

Towards black-box Koopmans band structures or: getting lost down a pseudopotential-generation rabbit hole



Koopmans functionals give accurate band structures



Mean absolute error (eV) across prototypical semiconductors and insulators

	PBE	G_0W_0	KI	KIPZ	$QSG\tilde{W}$
E_{gap}	2.54	0.56	0.27	0.22	0.18
IP	1.09	0.39	0.19	0.21	0.49

	PBE	$G_0W_0^1$	scGW ²	KI@[PBE,MLWFs]	KIPZ@PBE	exp ³
E_g	0.49	1.06	1.14	1.16	1.15	1.17
$\Gamma_{1v} \rightarrow \Gamma_{25'v}$	11.97	12.04		11.97	12.09	12.5 ± 0.6
$X_{1v} \rightarrow \Gamma_{25'v}$	7.82			7.82		7.75
$X_{4v} \rightarrow \Gamma_{25'v}$	2.85	2.99		2.85	2.86	2.90
$L_{2'v} \rightarrow \Gamma_{25'v}$	9.63	9.79		9.63	9.74	9.3 ± 0.4
$L_{1v} \rightarrow \Gamma_{25'v}$	6.98	7.18		6.98	7.04	6.8 ± 0.2
$L_{3'v} \rightarrow \Gamma_{25'v}$	1.19	1.27		1.19		1.2 ± 0.2
$\Gamma_{25'v} \rightarrow \Gamma_{15c}$	2.48	3.29		3.17	3.20	3.35 ± 0.01
$\Gamma_{25'v} \rightarrow \Gamma_{2'c}$	3.28	4.02		3.95	3.95	4.15 ± 0.05
$\Gamma_{25'v} \rightarrow X_{1c}$	0.62	1.38		1.28	1.31	1.13
$\Gamma_{25'v} \rightarrow L_{1c}$	1.45	2.21		2.12	2.13	2.04 ± 0.06
$\Gamma_{25'v} \rightarrow L_{3c}$	3.24	4.18		3.91	3.94	3.9 ± 0.1
MSE	0.35	0.02		0.01	0.03	
MAE	0.44	0.21		0.14	0.17	

¹ M. Shishkin et al. *Phys. Rev. Lett.* 99.24 (2007), 246403 for E_g and M. S. Hybertsen et al. *Phys. Rev. B* 34.8 (1986), 5390 for the transitions;

² M. Shishkin et al. *Phys. Rev. B* 75.23 (2007), 235102.

³ O. Madelung. *Semiconductors*. 3rd ed. Berlin: Springer-Verlag, 2004.

Features of Koopmans functionals

$$E_{\text{Koopmans}}[\rho, \{f_i\}, \{\alpha_i\}] = E_{DFT}[\rho] + \sum_i \alpha_i \left(- \int_0^{f_i} \varepsilon_i(f) df + f_i \eta_i \right)$$

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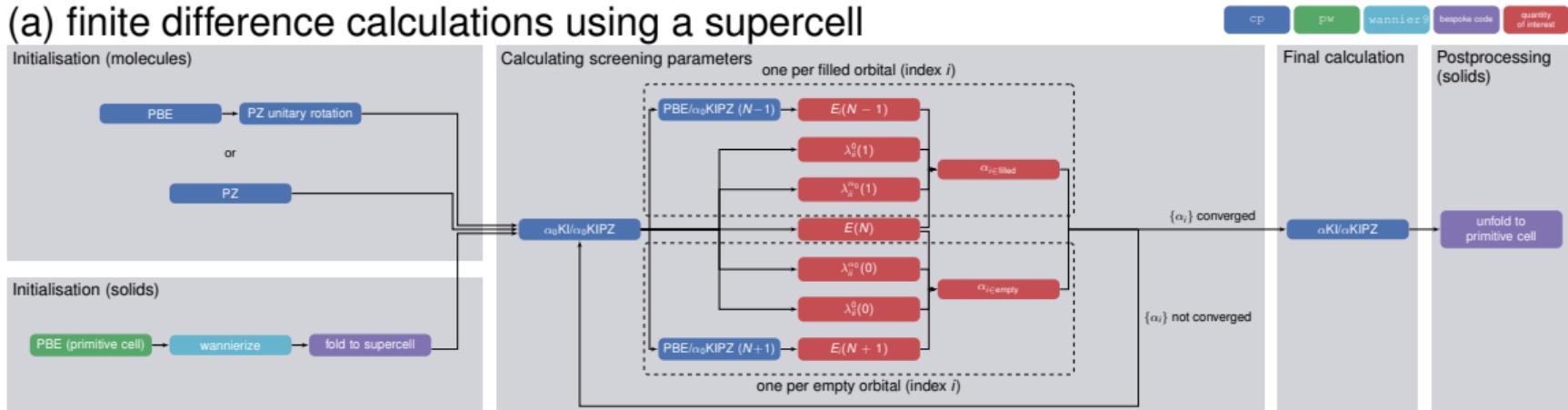
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In order to evaluate this functional, one must...

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- calculate the screening parameters $\{\alpha_i\}$
- construct and diagonalize the Hamiltonian

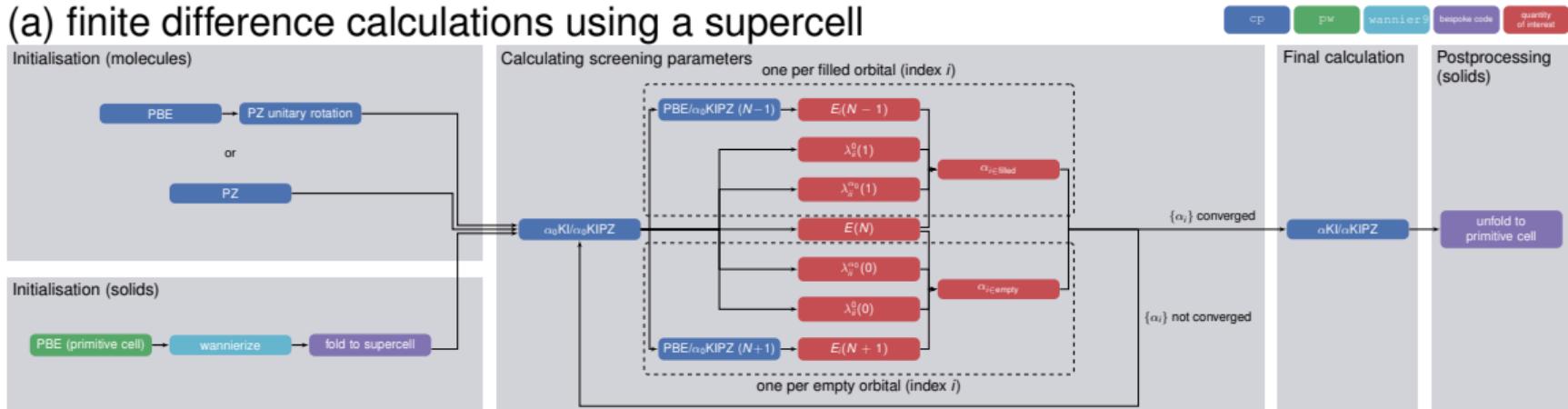
Workflows

(a) finite difference calculations using a supercell

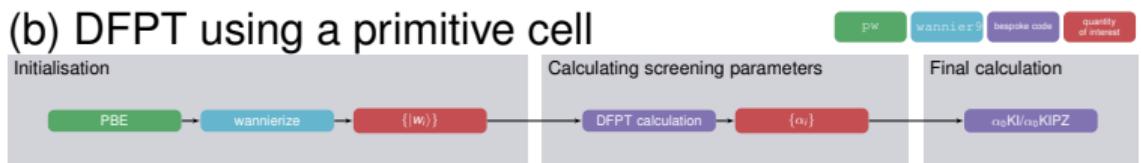


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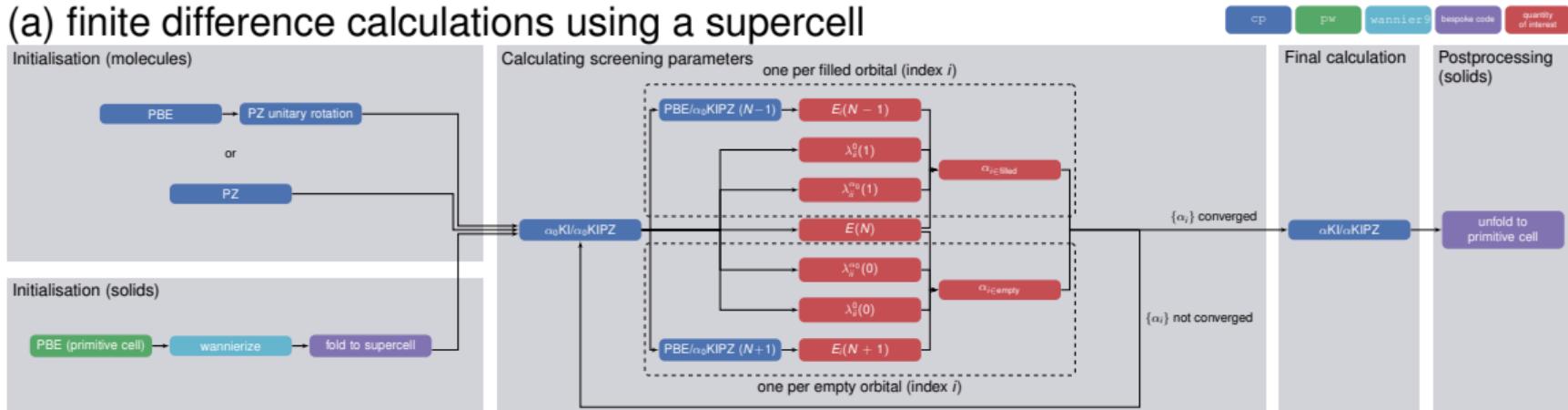


(b) DFPT using a primitive cell

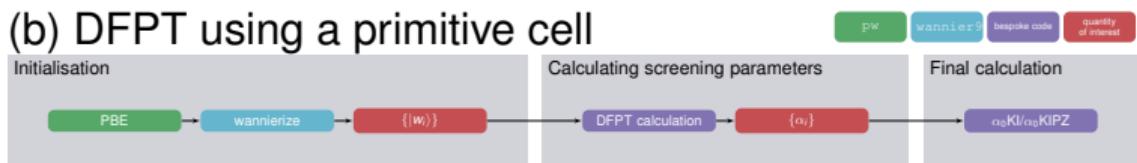


Workflows

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(b) DFPT using a primitive cell



All implemented in Koopmans

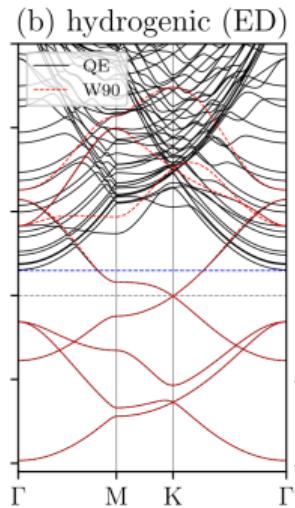
Workflows

What still stands in our way? Take the example of silicon:

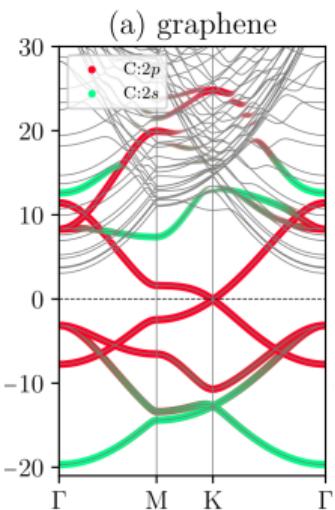
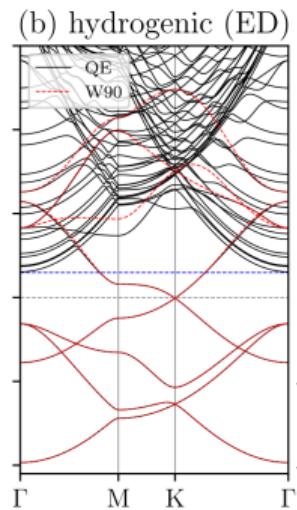
```
{  
  "workflow": {  
    "task": "singlepoint",  
    "functional": "ki",  
    "base_functional": "lda",  
    "method": "dfpt",  
    "pseudo_library": "pseudo_dojo_standard"},  
  "atoms": {  
    "cell_parameters": {"periodic": true, "ibrav": 2, "celldms": {"1": 10.2622}},  
    "atomic_positions": {  
      "units": "crystal",  
      "positions": [[{"Si": 0.00, 0.00, 0.00}, {"Si": 0.25, 0.25, 0.25}]]},  
    "kpoints": {"grid": [8, 8, 8]},  
    "calculator_parameters": {  
      "ecutwfc": 60.0,  
      "pw": {"nbnd": 20},  
      "w90": {  
        "projections": [[[{"fsite": [0.25, 0.25, 0.25], "ang_mtm": "sp3"}],  
                      [{"fsite": [0.25, 0.25, 0.25], "ang_mtm": "sp3"}]]],  
        "dis_froz_max": 10.6,  
        "dis_win_max": 16.9}}}}
```

One very manual step: Wannierization. Can we automate this?

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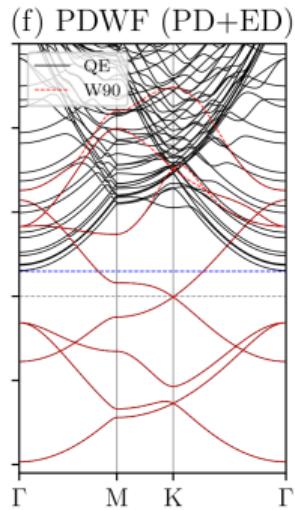
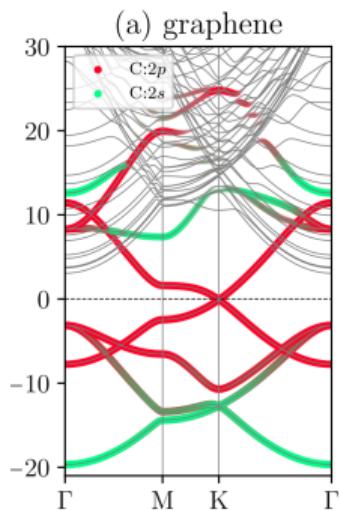
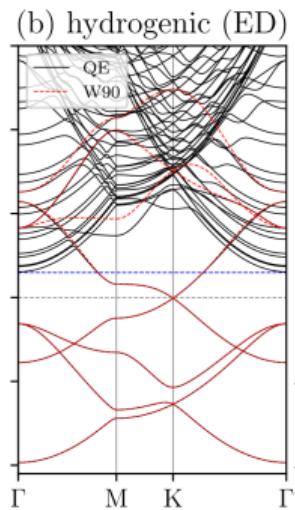


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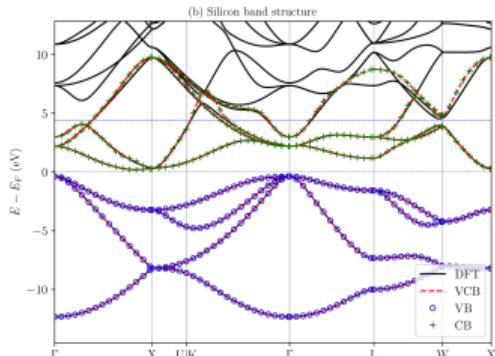


Automating Wannierization

One very manual step: Wannierization. Can we automate this?



We separate target manifolds via parallel transport to obtain separate occupied and empty manifolds



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- we use them to calculate projectability
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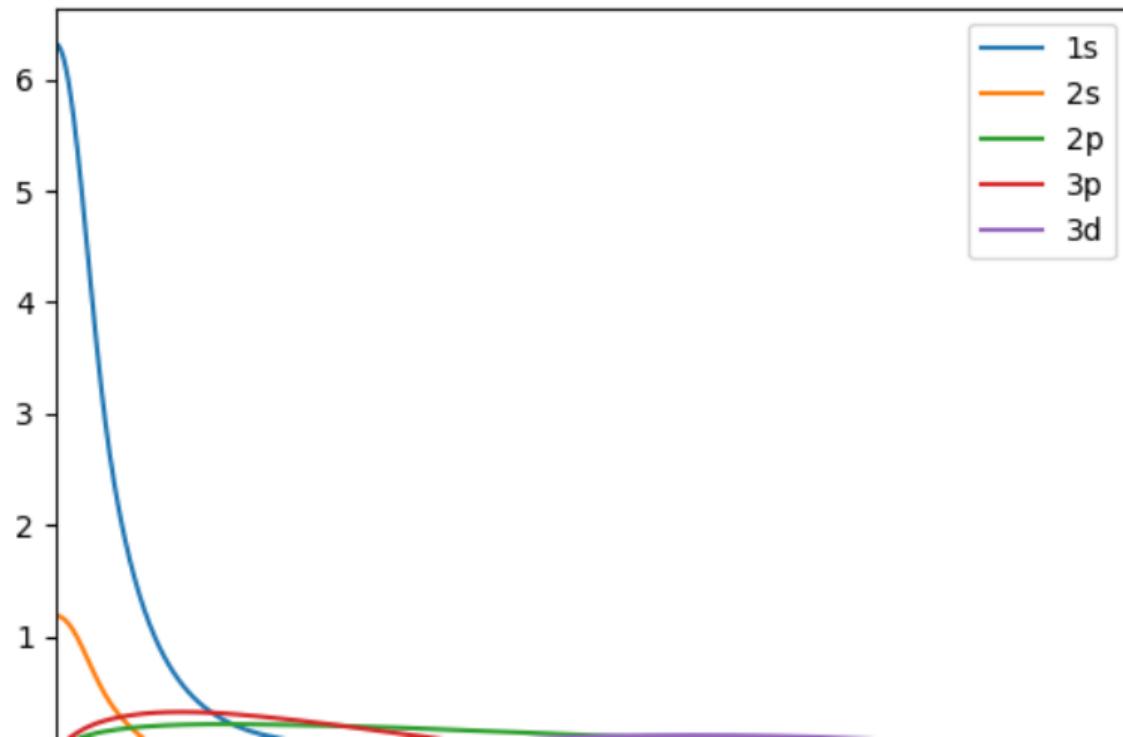
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6 Wannier functions for a system with 10 electrons = 5 occupied bands + only 1 unoccupied band!

If we want more Wannier functions, we're gonna need ~~a bigger boat~~ more PAOs...

Automating Wannierization

Existing strategy: use the PAOs provided by OpenMX



The screenshot shows a GitHub browser interface for the `upf-tools` repository. The URL in the address bar is `github.com/elinscott/upf-tools/blob/main/README.md`. The page displays the `README.md` file content.

File Tree:

- `main`
- `.github`
- `docs`
- `src`
- `tests`
- `.bumpversion.cfg`
- `.gitignore`
- `.readthedocs.yml`
- `CITATION.cff`
- `LICENSE`
- `MANIFEST.in`
- `README.md` (selected)
- `pyproject.toml`
- `setup.cfg`
- `tox.ini`

README.md Content:

upf-tools

Tests passing | pypi v0.1.1 | python 3.8 | 3.9 | 3.10 | 3.11 | license MIT | docs passing | codecov 79% | Cookiecutter | snekpack | code style black | Contributor Covenant 2.1

Tools for handling `.upf` (Unified Pseudopotential Format) files

Getting Started

```
from upf_tools import UPFDict
psp = UPFDict.from_upf('/path/to/file.upf')
```

`UPFDict` is a lightweight class that behaves like a dictionary with a few added functionalities.

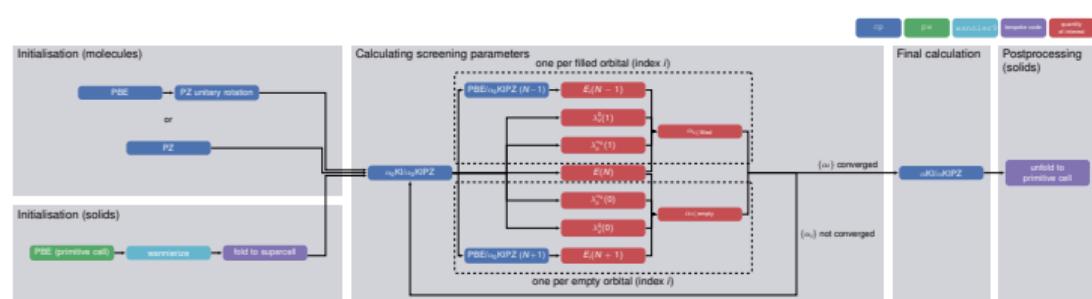
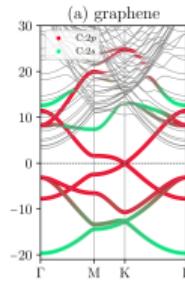
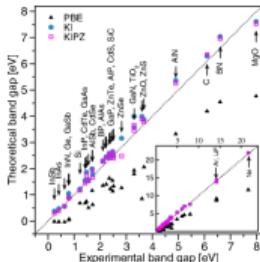
Command Line Interface

The `upf_tools` command line tool is automatically installed. It can be used from the shell with the `--help`

Most of the infrastructure was from the `cookiecutter` I used (see my July 2022 GM)
Already used in `AiiDA`

Also have unmerged tools for `oncvpsp` input and output files as well as Junfeng's custom
.dat projector files (perhaps `oncvpsp-tools`) Need to decide on the right home for
these

Take home messages



- Koopmans functionals yield band structures with comparable accuracy to state-of-the-art GW
- the release of `koopmans` means non-experts can now use Koopmans functionals in their own research
- work is ongoing to automate the Wannierization bottleneck

Acknowledgements



Nicola Marzari



Nicola Colonna



Riccardo De Gennaro

Junfeng Qiao



**Swiss National
Science Foundation**

MARVEL
The logo for MARVEL consists of four red hexagons arranged in a horizontal row, with the second and third hexagons partially overlapping.

NATIONAL CENTRE OF COMPETENCE IN RESEARCH

slides available at  [github/elinscott-talks](https://github.com/elinscott-talks) and on the THEOS wiki