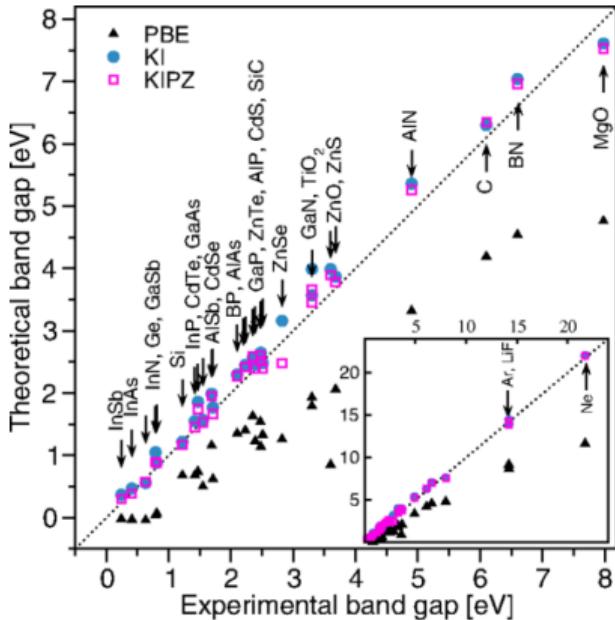


# Automating the calculation of accurate band structures with Koopmans functionals

# KOOPMANS FUNCTIONALS GIVE ACCURATE BAND STRUCTURES



Mean absolute error (eV) across prototypical semiconductors and insulators

	PBE	G <sub>0</sub> W <sub>0</sub>	KI	KIPZ	QSGW
$E_{\text{gap}}$	2.54	0.56	0.27	0.22	0.18
IP	1.09	0.39	0.19	0.21	0.49

# KOOPMANS FUNCTIONALS GIVE ACCURATE BAND STRUCTURES

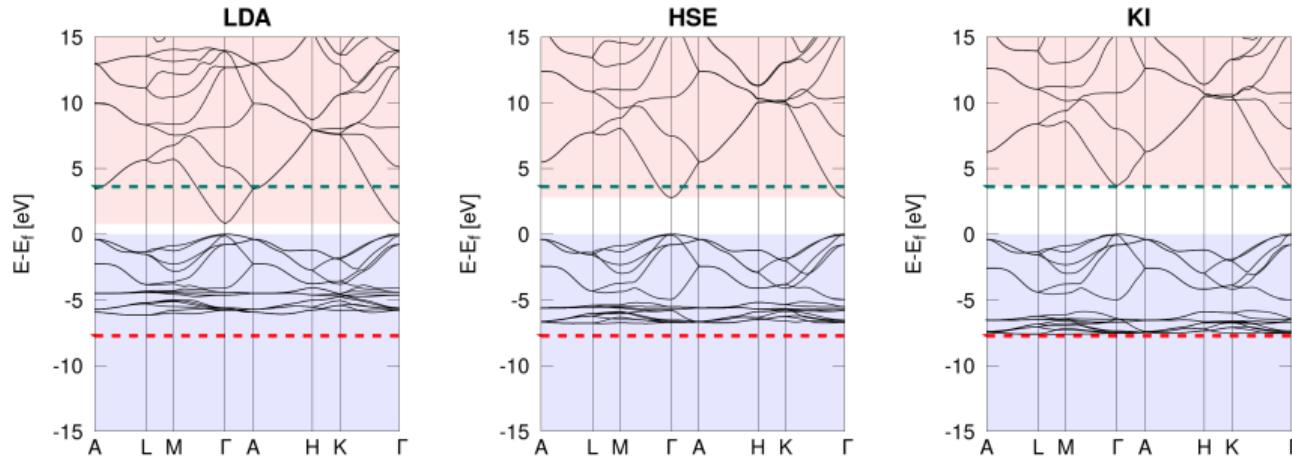
	PBE	$G_0W_0^1$	scGW $\tilde{W}^2$	KI@[PBE,MLWFs]	KIPZ@PBE	exp $^3$
$E_g$	0.49	1.06	1.14	1.16	1.15	1.17
$\Gamma_{1v} \rightarrow \Gamma_{25'v}$	11.97	12.04		11.97	12.09	12.5 $\pm$ 0.6
$X_{1v} \rightarrow \Gamma_{25'v}$	7.82			7.82		7.75
$X_{4v} \rightarrow \Gamma_{25'v}$	2.85	2.99		2.85	2.86	2.90
$L_{2'v}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

<sup>1</sup> M. Shishkin et al. *Phys. Rev. B* 75.23 (2007), 235102 for  $E_g$  and M. S. Hybertsen et al. *Phys. Rev. B* 34.8 (1986), 5390 for the transitions;

<sup>2</sup> M. Shishkin et al. *Phys. Rev. Lett.* 99.24 (2007), 246403.

<sup>3</sup> O. Madelung. *Semiconductors*. 3rd ed. Berlin: Springer-Verlag, 2004.

# KOOPMANS FUNCTIONALS GIVE ACCURATE BAND STRUCTURES



ZnO	LDA	HSE	$GW_0$	$scG\tilde{W}$	KI	exp
$E_{gap}$ (eV)	0.79	2.79	3.0	3.2	3.62	3.60
$\langle \varepsilon_d \rangle$ (eV)	-5.1	-6.1	-6.4	-6.7	-6.9	-7.5/-8.0

N. Colonna et al. *J. Chem. Theory Comput.* 18.9 (2022), 5435

# FEATURES OF KOOPMANS FUNCTIONALS

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

General features:

- enforces a generalized piecewise linearity condition
- depends on localization
- is orbital-density-dependent
- requires the ab initio calculation of screening parameters

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

- define/initialize a set of variational orbitals
- calculate the screening parameters  $\{\alpha_i\}$
- construct and diagonalize the Hamiltonian

# WORKFLOWS

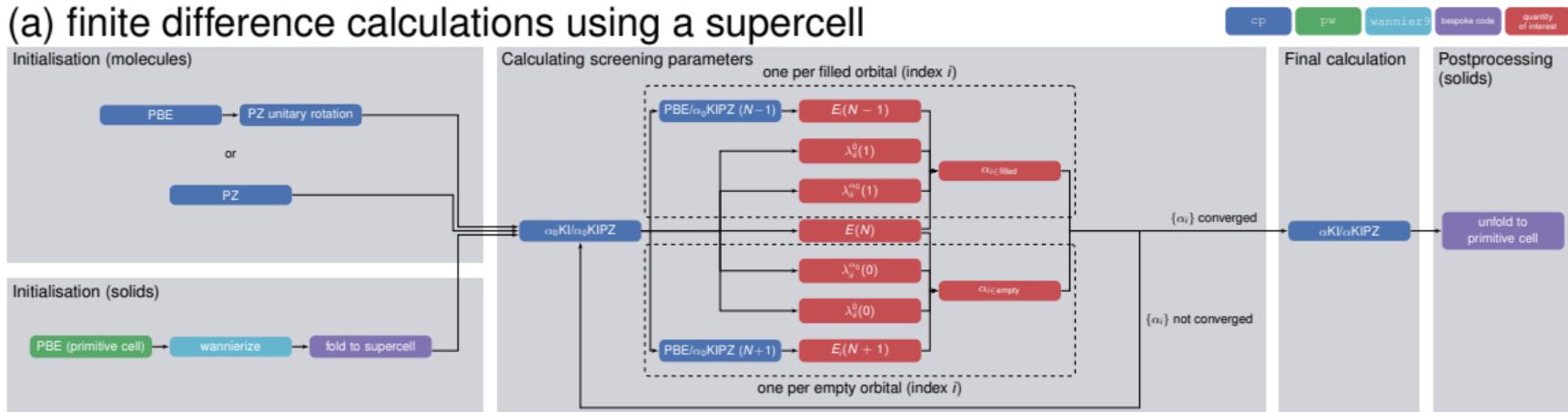
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E. B. Linscott et al. *J. Chem. Theory Comput.* (2023)



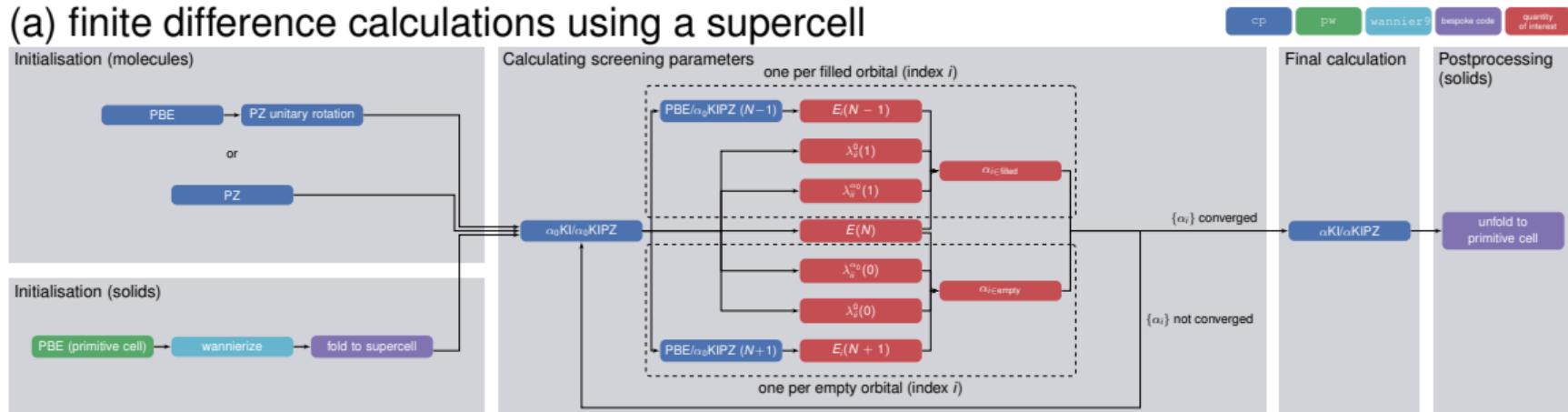
# WORKFLOWS

## (a) finite difference calculations using a supercell

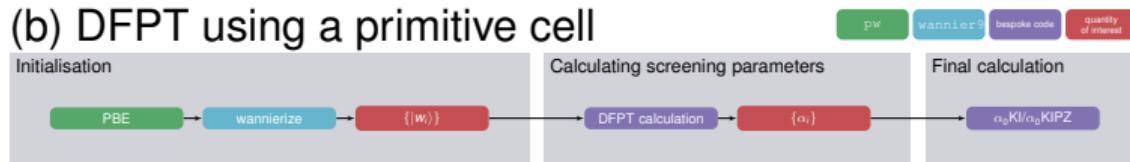


# WORKFLOWS

## (a) finite difference calculations using a supercell



## (b) DFPT using a primitive cell



# WORKFLOWS

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Complicated workflows mean that...

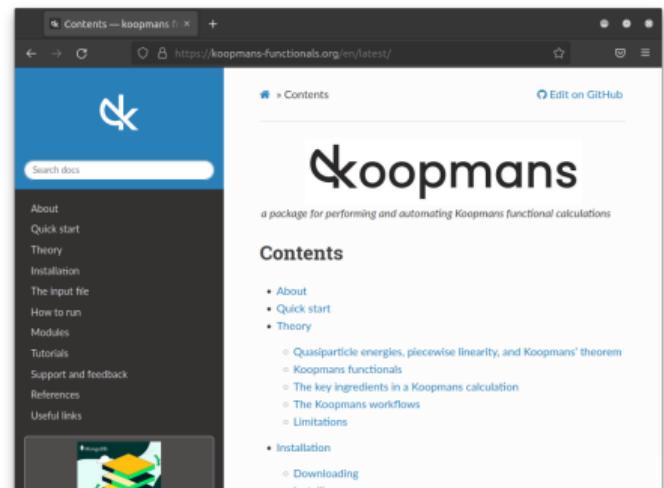
- lots of different codes that need to handshake
- lots of scope for human error
- reproducibility becomes difficult
- expert knowledge required

Our solution...

# koopmans

- v1.0 released earlier this year
- implementations of Koopmans functionals within Quantum ESPRESSO
- automated workflows
  - start-to-finish Koopmans calculations
  - Wannierisation
  - dielectric tensor
  - convergence tests
  - ...
- built on top of ASE
- does not require expert knowledge

[koopmans-functionals.org](https://koopmans-functionals.org)



# koopmans: An Open-Source Package for Accurately and Efficiently Predicting Spectral Properties with Koopmans Functionals

Edward B. Linscott,\*<sup>△</sup> Nicola Colonna,<sup>△</sup> Riccardo De Gennaro, Ngoc Linh Nguyen, Giovanni Borghi, Andrea Ferretti, Ismaila Dabo, and Nicola Marzari\*



Cite This: <https://doi.org/10.1021/acs.jctc.3c00652>



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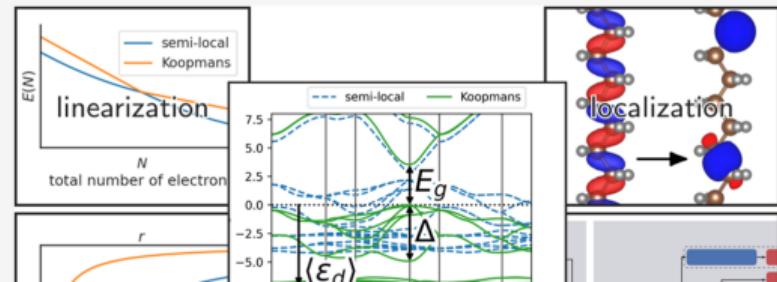
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**ABSTRACT:** Over the past decade we have developed Koopmans functionals, a computationally efficient approach for predicting spectral properties with an orbital-density-dependent functional framework. These functionals impose a generalized piecewise linearity condition to the entire electronic manifold, ensuring that orbital energies match the corresponding electron removal/addition energy differences (in contrast to semilocal DFT, where a mismatch between the two lies at the heart of the band gap



# AUTOMATING WANNIERIZATION

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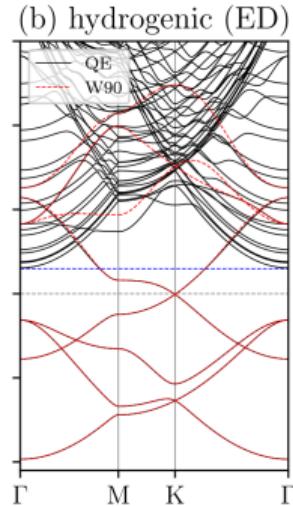
One remaining manual step: Wannierization. Can we automate this?

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J. Qiao et al. *Projectability Disentanglement for Accurate and Automated Electronic-Structure Hamiltonians*.  
<http://arxiv.org/abs/2303.07877>. 2023

# AUTOMATING WANNIERIZATION

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

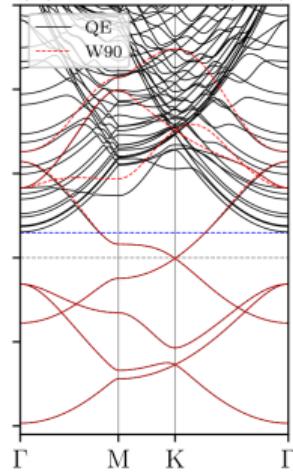


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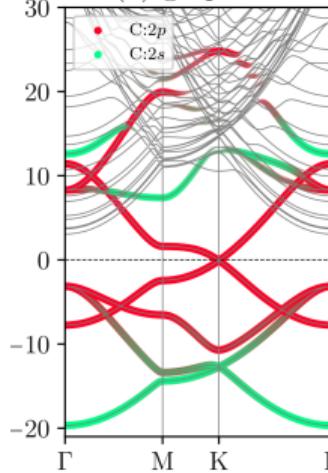
# AUTOMATING WANNIERIZATION

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

(b) hydrogenic (ED)



(a) graphene

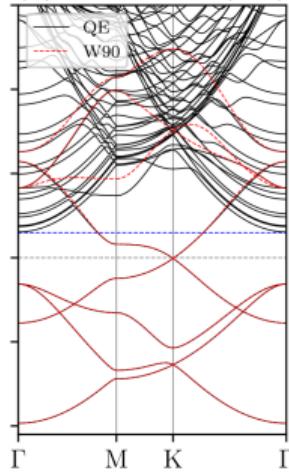


J. Qiao et al. *Projectability Disentanglement for Accurate and Automated Electronic-Structure Hamiltonians*.  
<http://arxiv.org/abs/2303.07877>. 2023

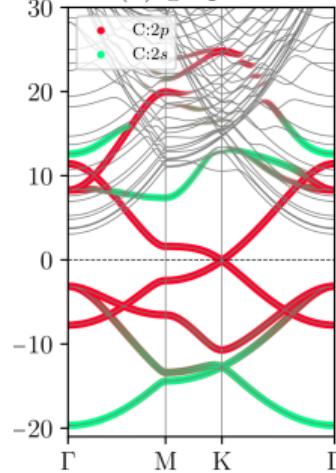
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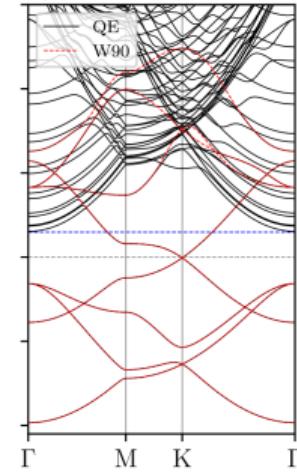
(b) hydrogenic (ED)



(a) graphene



(f) PDWF (PD+ED)

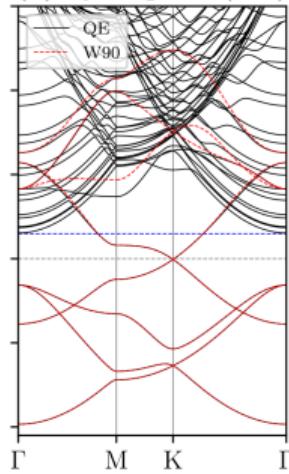


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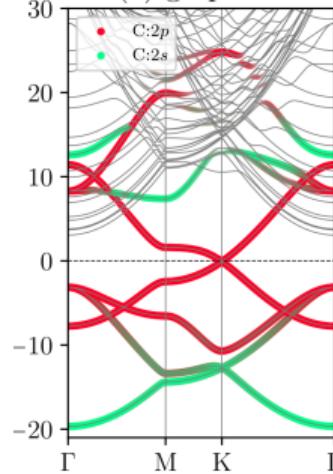
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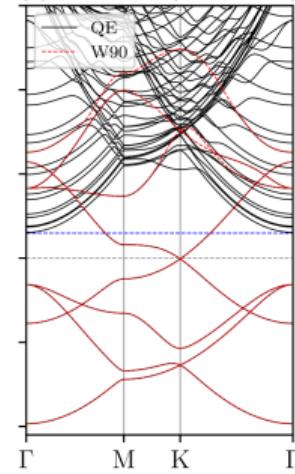
(b) hydrogenic (ED)



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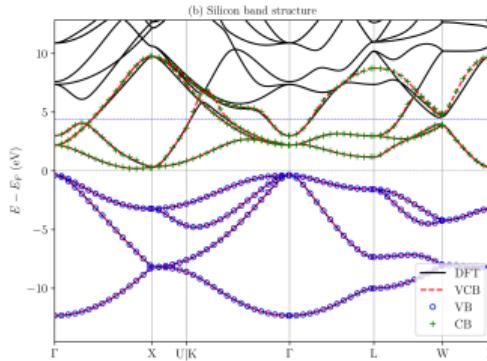
Demonstrated on >20,000 materials → black-box Wannierization

J. Qiao et al. *Projectability Disentanglement for Accurate and Automated Electronic-Structure Hamiltonians*.

<http://arxiv.org/abs/2303.07877>. 2023

# AUTOMATING WANNIERIZATION

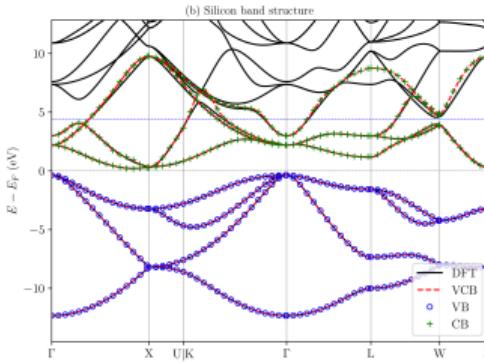
We separate target manifolds via parallel transport to obtain separate occupied and empty manifolds



J. Qiao et al. *Automated Mixing of Maximally Localized Wannier Functions into Target Manifolds*.  
<http://arxiv.org/abs/2306.00678>. 2023

# AUTOMATING WANNIERIZATION

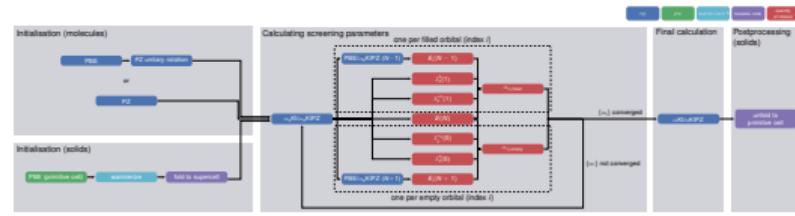
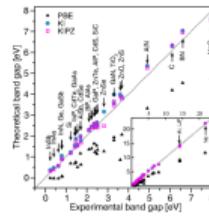
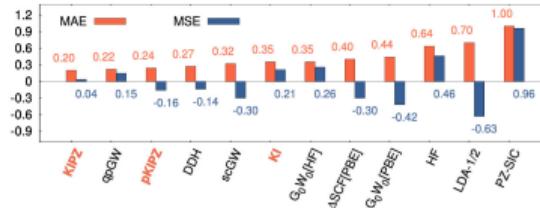
We separate target manifolds via parallel transport to obtain separate occupied and empty manifolds



All of the above is supported by koopmans, but more details to be resolved... watch this space!

J. Qiao et al. *Automated Mixing of Maximally Localized Wannier Functions into Target Manifolds*.  
<http://arxiv.org/abs/2306.00678>. 2023

# TAKE HOME MESSAGES



- Koopmans functionals can give orbital energies and 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

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Nicola Marzari



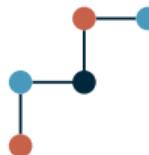
Nicola Colonna



Riccardo De Gennaro



Yannick Schubert



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