First principles simulation of Quantum Materials

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SURF Advanced Computing User Day 2024

Materials for the Quantum Age

Overview

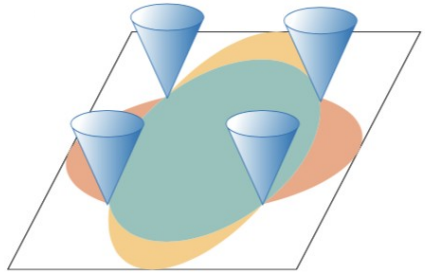
1) Quantum Materials

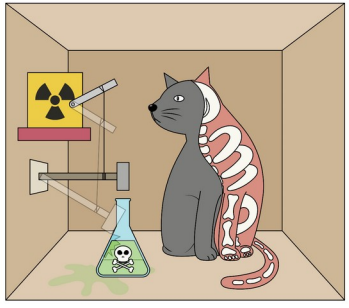
2) First-principles simulations

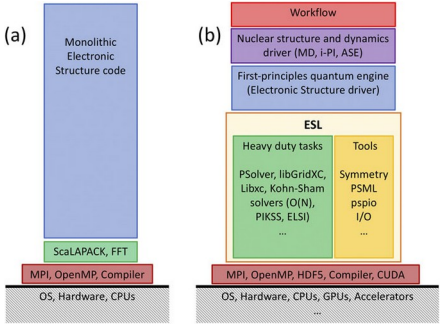
3) High-performance computing

4) Good practices

5) Future







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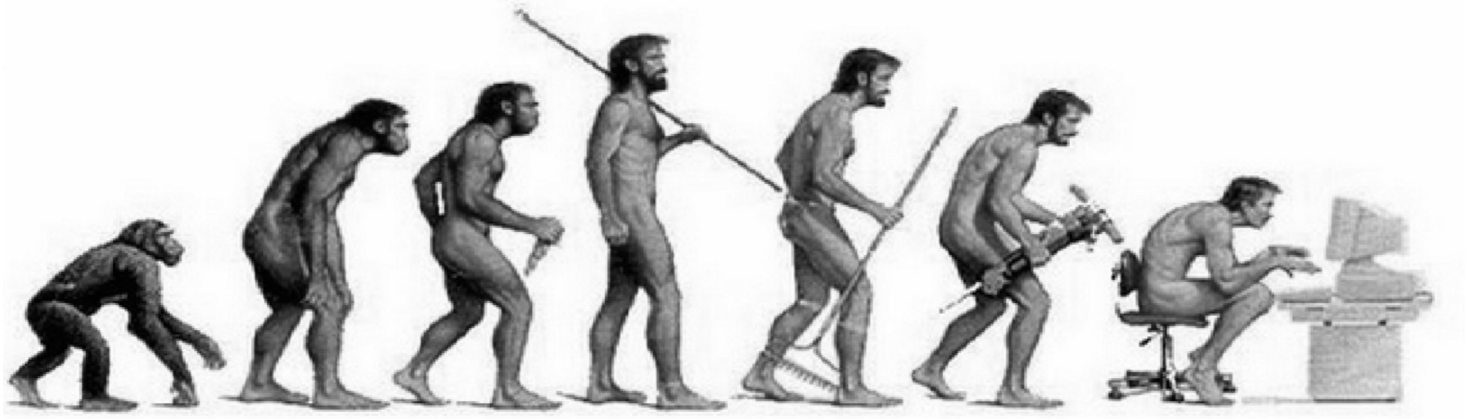
Quantum Materials

What drives human development?

Major advances in human civilization are driven by materials:

stone, bronze, iron… **… silicon age**



But... **The silicon era is approaching its limits.**

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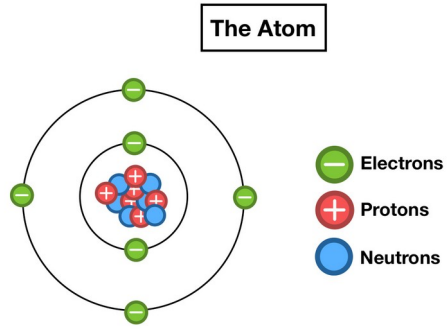
Quantum Materials



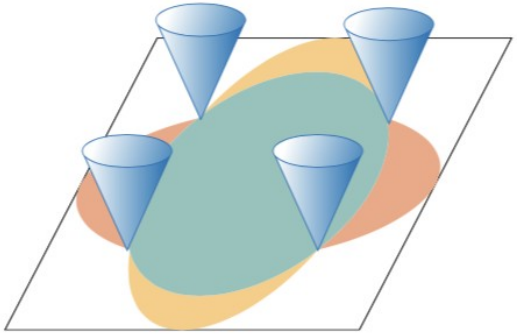
All materials are quantum

But some materials are more quantum than others.

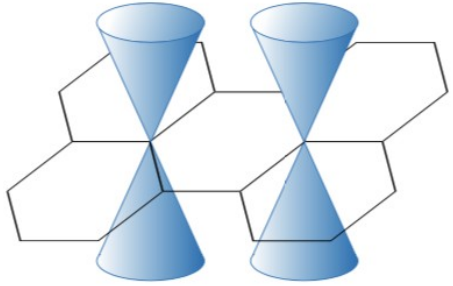
- Zeila Zanolli (2020)

Quantum Materials

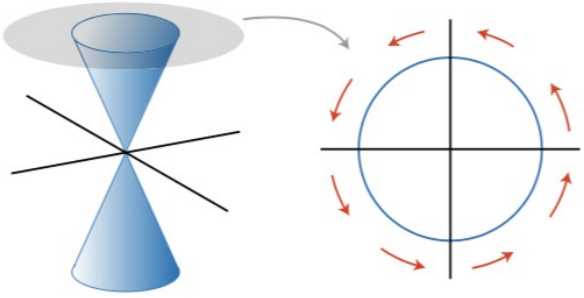
Superconductors



Dirac Materials



Topological insulators



Keimer & Moore Nat Phys 2017

Quantum behaviour at macroscopic scales

Quantum technology:
Low-dissipation electronics
Photovoltaics
Quantum information

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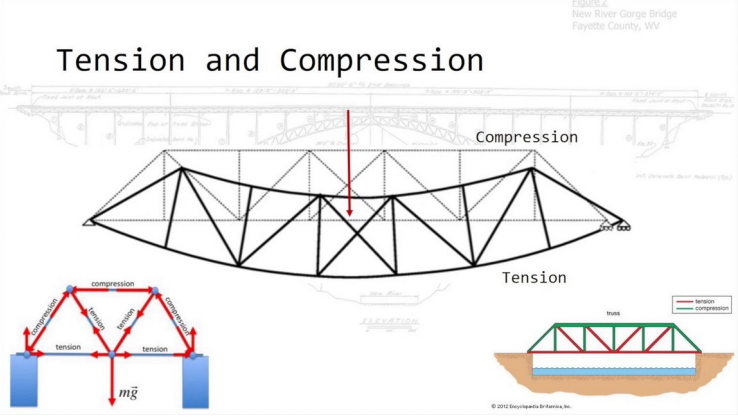
Quantum Materials by Design group

Using simulations to guide quantum materials design

- Perfect atomic control
- Try before you buy/fabricate
- Easy exploration of many different materials/combinations
- Close collaboration with experimentalists



He tested thousands and thousands of other materials to use for the filament.

The Franklin Institute on Thomas Edison's search for the lightbulb filament *https://fi.edu/en/science-and-education/collection/edisons-lightbulb*

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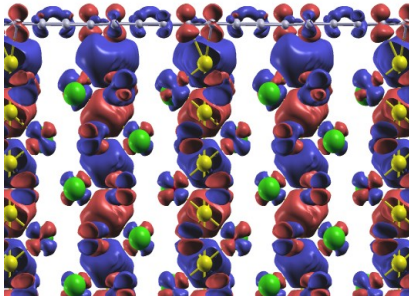
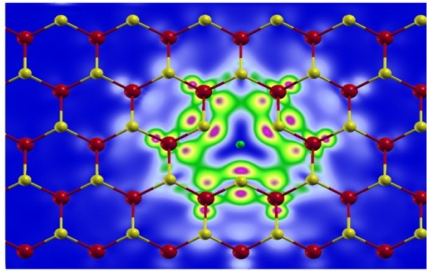
Quantum Materials for Quantum Technologies & Sustainability



Topological Matter

- Low-energy consumption electronics



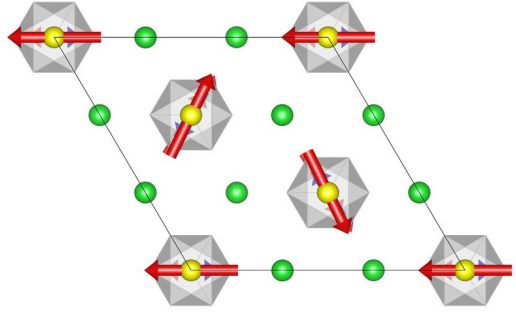
Theoretical Spectroscopy

- Quantum computing
- Cryptography
- Quantum sensing
- Photovoltaics

Quantum Materials

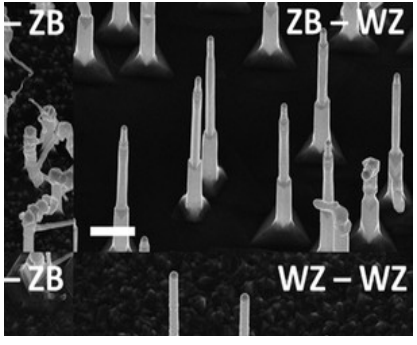
Magnetism

- Spin Qubits



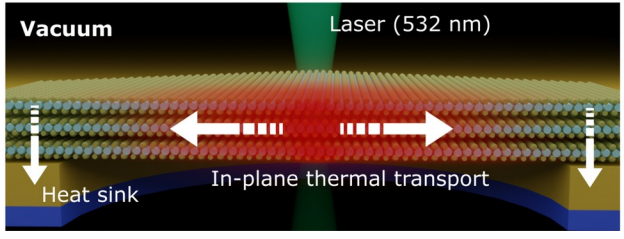
Superconductivity

- Q computing, Qubits



Quantum Electron & Thermal Transport

- Semiconductor industry



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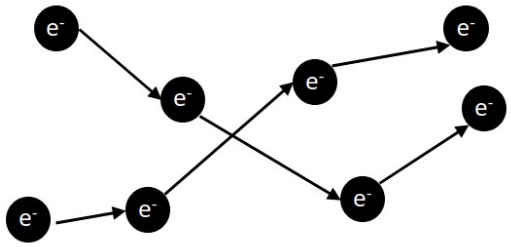
Normal metals vs superconductors

Normal metal



Electrons repel each other (Coulomb)

Electron-electron scattering
→ finite resistance

Superconductor

Effective attractive electron-electron interaction
- ➔ Formation of Cooper pairs
- ➔ Collective motion without scattering

Normal metals vs superconductors

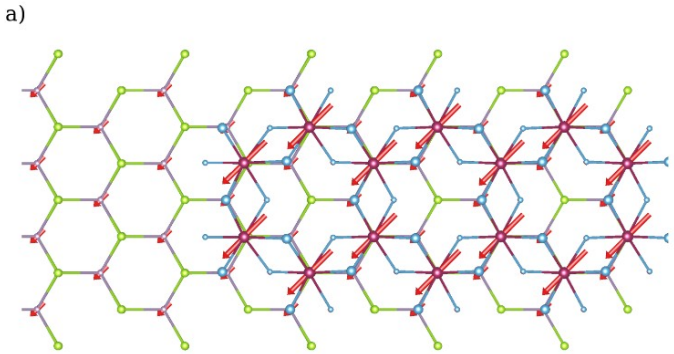
Superconductor

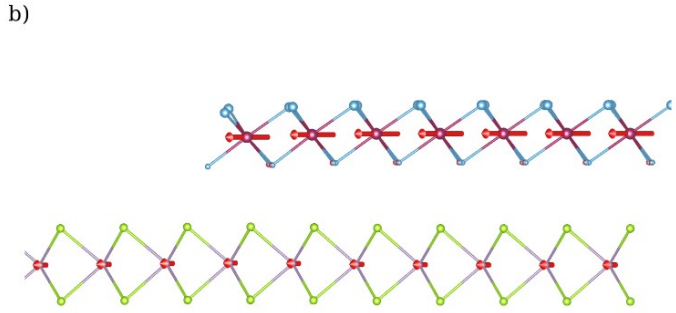
Effective attractive electron-electron interaction

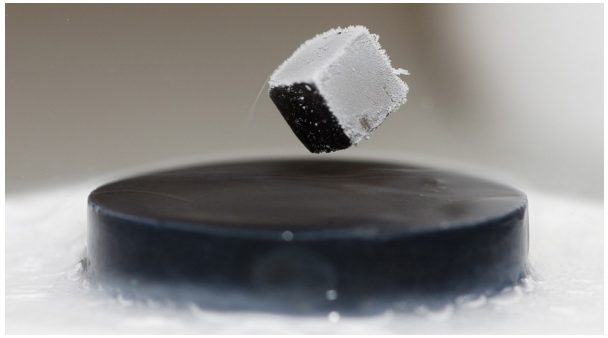
- ➔ Formation of Cooper pairs
- ➔ Collective motion without scattering

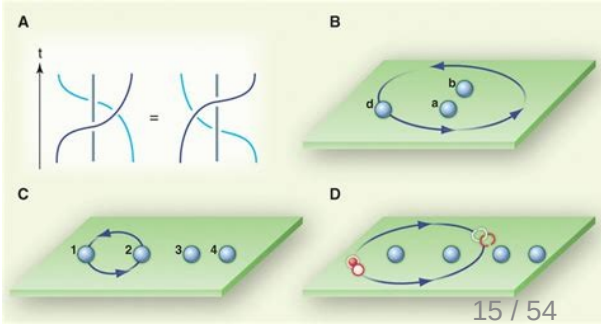
Interplay superconductivity and magnetism

- Conventional superconductors expel magnetic fields (Meissner effect)
- Heterostructures of magnetic atoms and superconductors yield exciting new physics
- Potential applications in quantum computing









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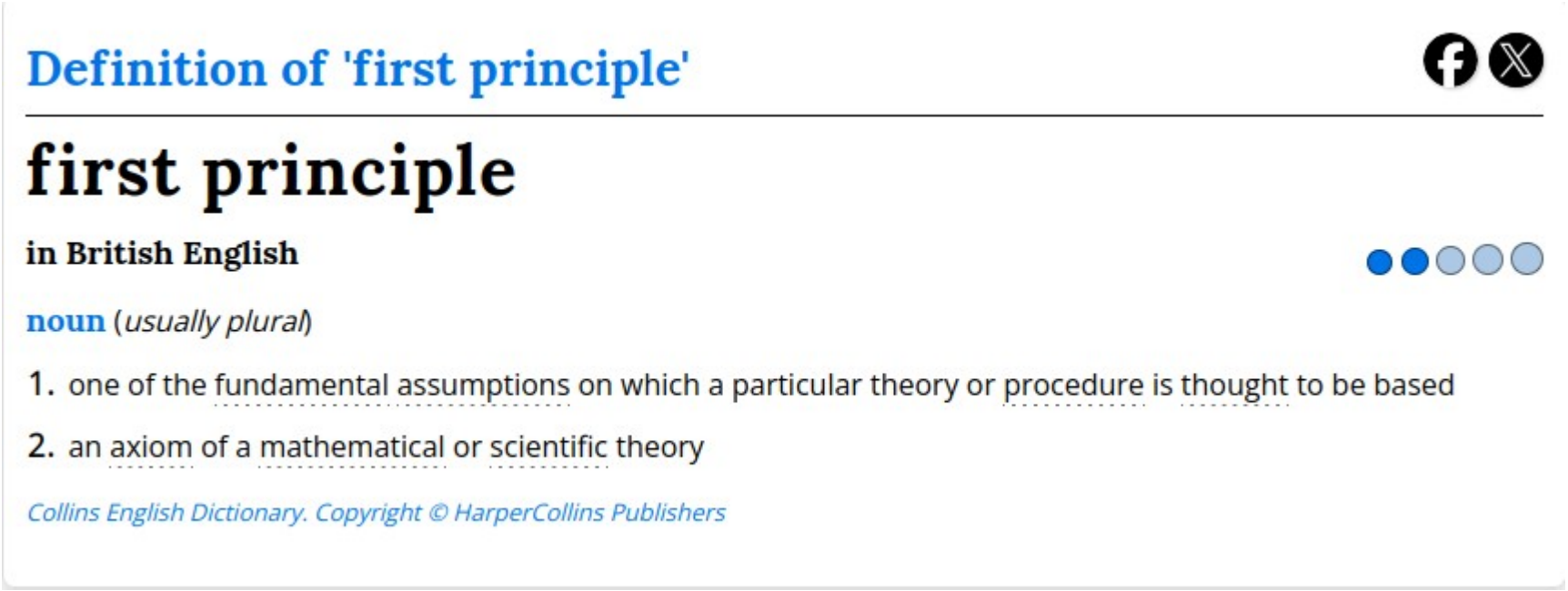
First-principles simulations

First-principles



Definition of 'first principle'

first principle

in British English

noun (*usually plural*)

1. one of the fundamental assumptions on which a particular theory or procedure is thought to be based
2. an axiom of a mathematical or scientific theory

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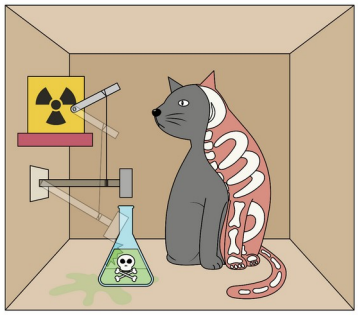
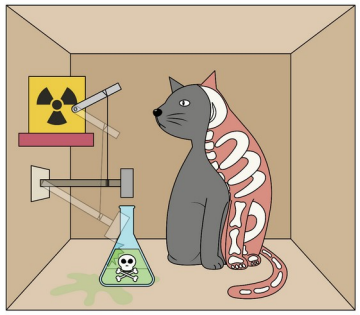
Computing the properties of the system in a **parameter-free** way, including all the material specific details

Quantum description



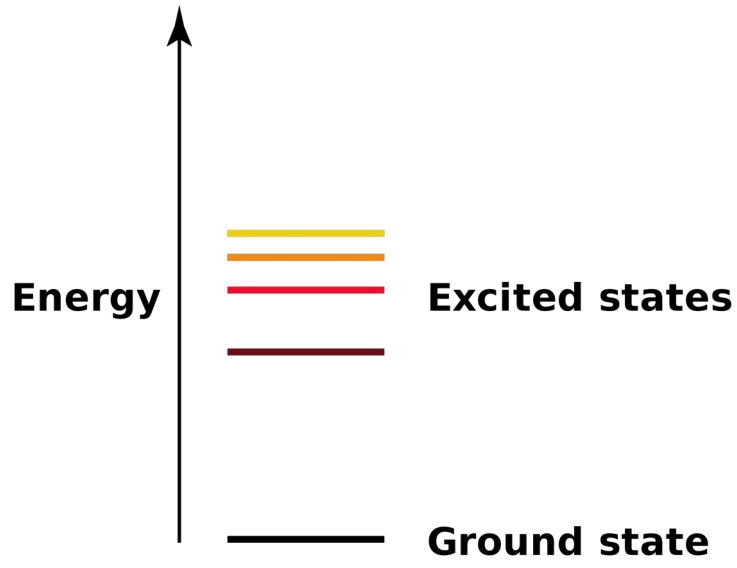
$$H\Psi_n = E_n\Psi_n$$

Quantum description



$$H\Psi_n = E_n\Psi_n$$

Solve



Quantized states Ψ_n with energy E_n

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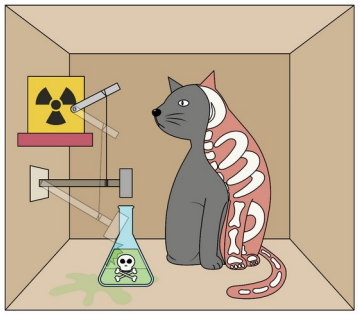
Quantum description



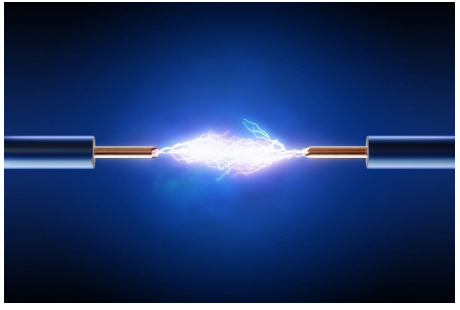
$$H\Psi_n = E_n\Psi_n$$

Solve

Ground state properties
- Electronic
- Structural
- Magnetic
- Topological

Excited state properties
- Optical properties

Time-dependent properties
- Transport



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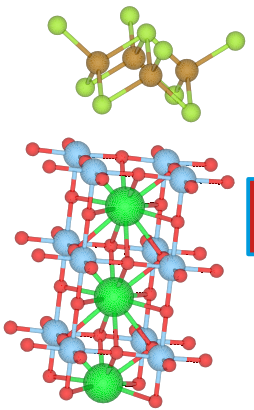
Overview of computational approach



Construct H

Solve for ground state

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Abinit

QUANTUMESPRESSO

Add SC

First-principles superconductivity

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SC: Superconductivity

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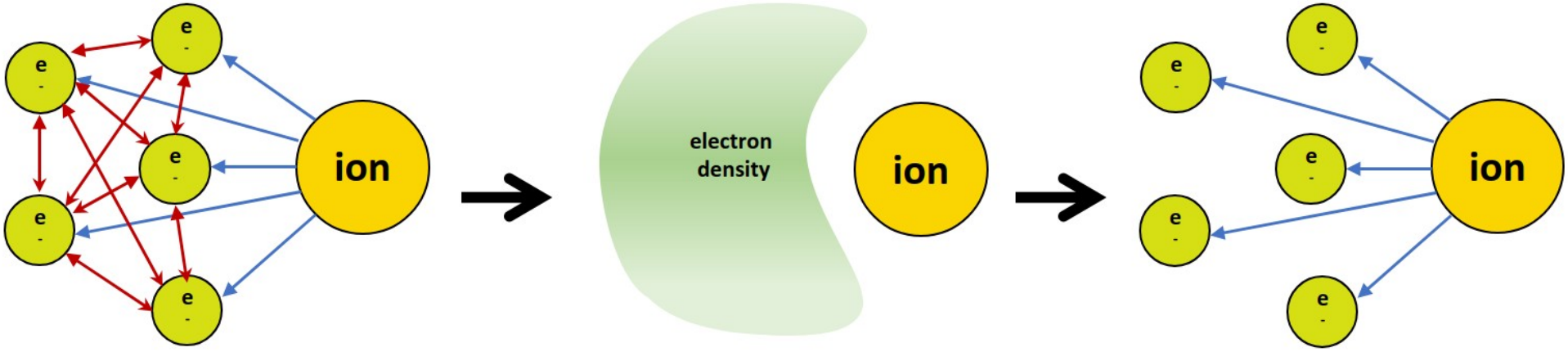
Density Functional Theory



Many-Body

N Interacting particles

$$\psi(r_1, s_1; r_2, s_2; \ldots; rN, sN)$$

$$H\psi = (T + V + U)\psi = E\psi$$

Hohenberg-Kohn

Energy is a unique functional of **ground state** electron density n

$$E = E[n(r)]$$

Kohn-Sham

N Independent particles auxiliary potential and same **ground state** electronic density

$$\sim\varphi_i(x, s)$$

$$H\,\varphi = \left(T - E_F + V_{eff}\right)\varphi = E\varphi$$

$$V_{eff}(\boldsymbol{x})[\rho] = V_{ext}(\boldsymbol{x}) + \int \frac{\rho(\boldsymbol{x}')}{|\boldsymbol{x} - \boldsymbol{x}'|}\, d\boldsymbol{x}' + \frac{\delta E_{xc}[\rho]}{\delta\rho(\boldsymbol{x})}$$

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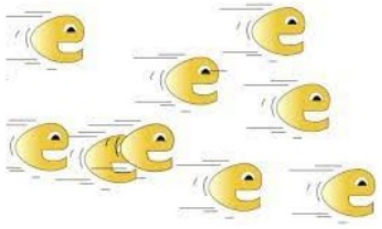
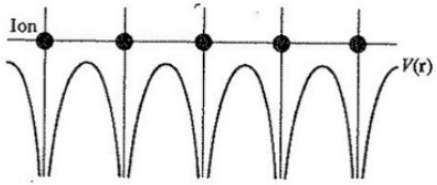
Hamiltonian

$$-\frac{1}{2}\nabla^2$$



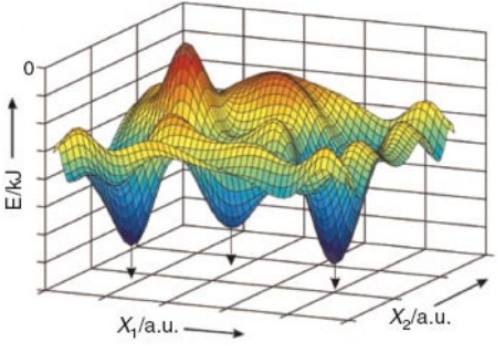
$$+$$

$$V_{\text{ext}}(\mathbf{r})$$



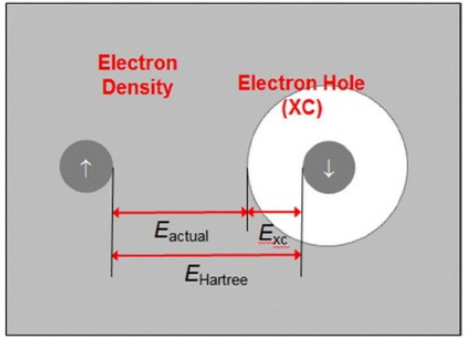
$$+$$

$$+$$



$$V_{\text{Hartree}}(\mathbf{r})$$

$$+$$



$$V_{\text{xc}}^{\sigma}(\mathbf{r})$$

Basis Sets

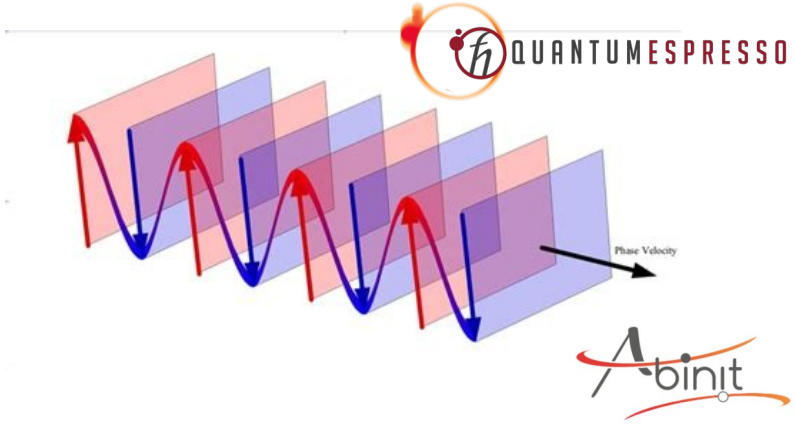
How to represent the wavefunction?

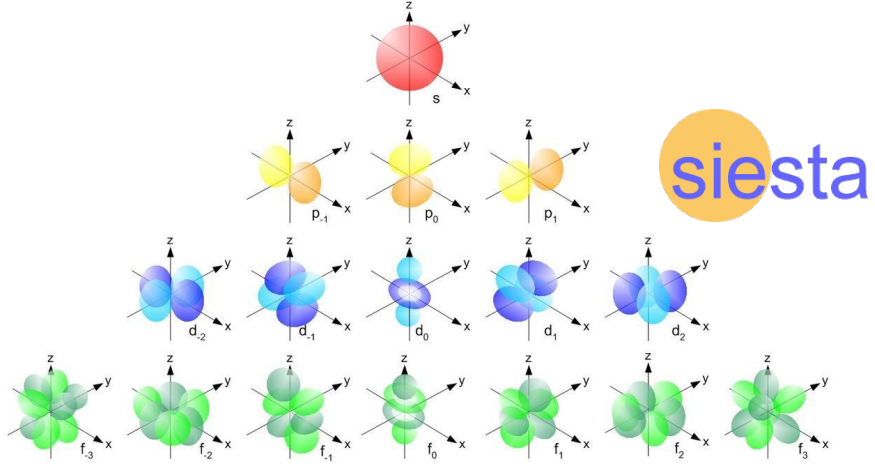
- On a regular grid => impractical
- Better: expand in terms of known functions

$$\psi = \sum_i c_i \phi_i$$

$$H_{ij} = \int \phi_i(r) H(r) \phi_j(r) dr$$

$$HC_i = E_i C_i$$





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First-principles superconductivity

$$H_{\mathrm{SC}} = H_{\mathrm{DFT}} + \Delta$$

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Our implementation in SIESTA

Reho, Wittemeier, Kole, Ordejón, Zanolli
Phys. Rev. B 110, 134505 (2024)

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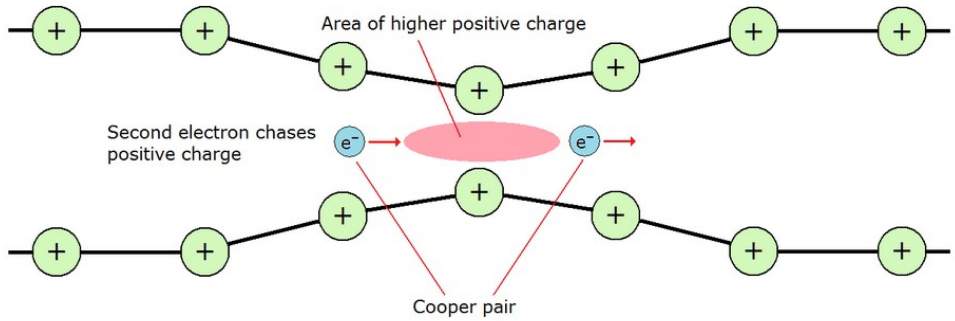

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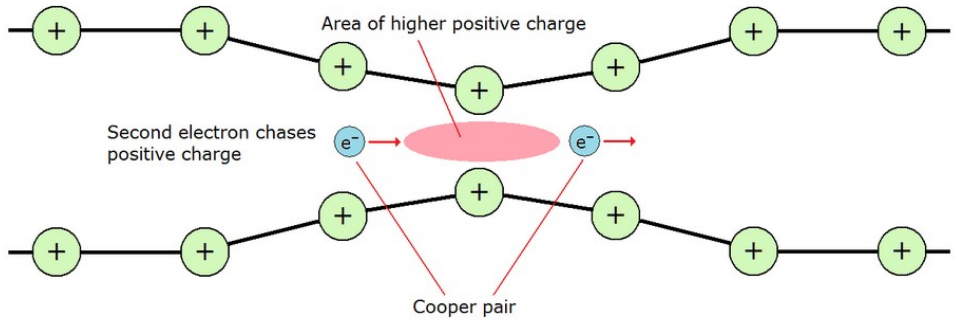

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$$\begin{pmatrix} H_{\mathrm{DFT}} & \Delta \\ -(\Delta)^* & -(H_{\mathrm{DFT}})^* \end{pmatrix} \begin{pmatrix} u_i^\sigma \\ v_i^\sigma \end{pmatrix} = \epsilon_i \begin{pmatrix} u_i^\sigma \\ v_i^\sigma \end{pmatrix}$$

u_i^σ electrons

v_i^σ holes

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First-principles superconductivity

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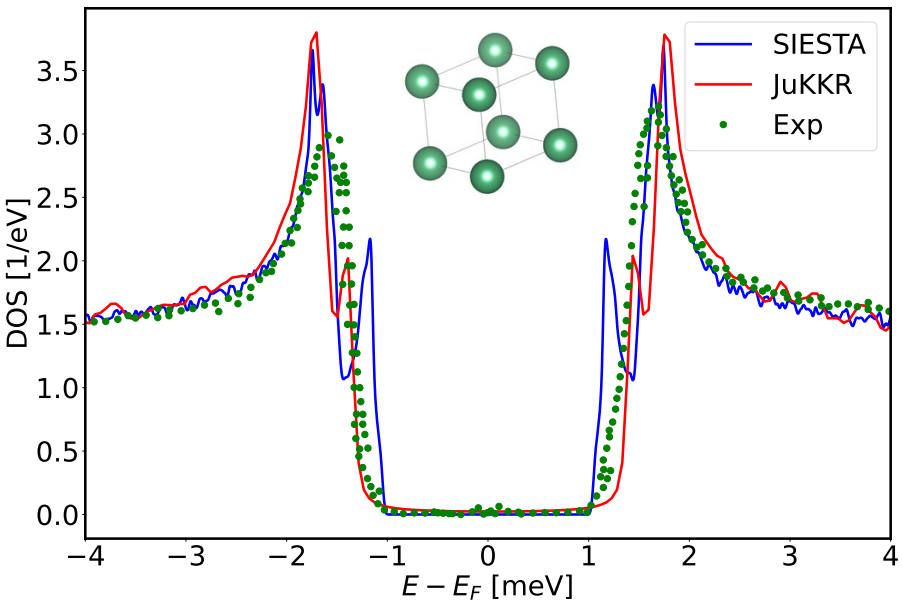

Reho, Wittemeier, Kole, Ordejón, Zanolli
Phys. Rev. B 110, 134505 (2024)

Example for bulk niobium

- Parameter-free except for superconducting interaction
- Direct comparison with experiment
- Measurable properties are material specific
- Small energy scales require high accuracy
- Valid for heterostructures

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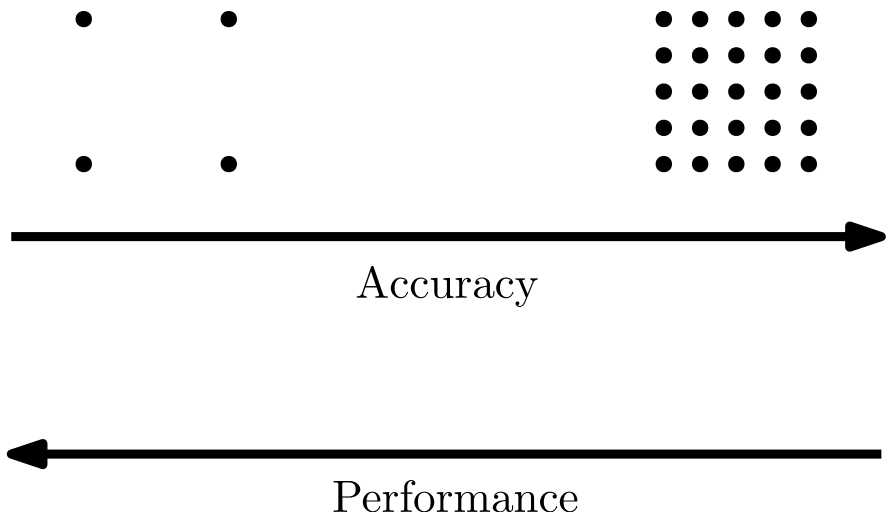
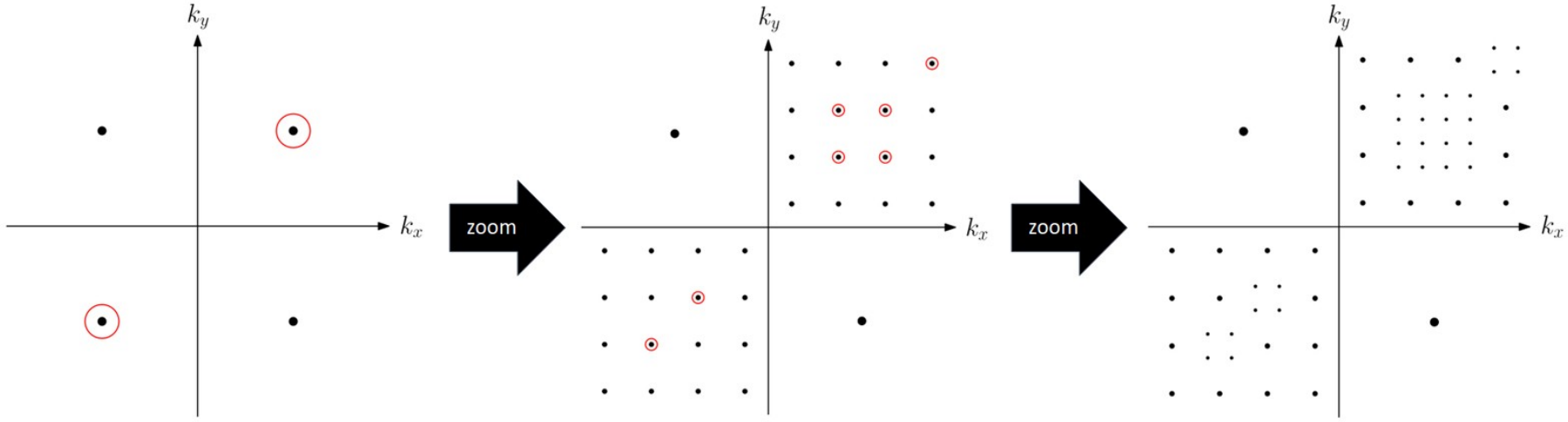
Performance vs Accuracy

- Size of basis set, number of grid points for integration, etc.
- Higher accuracy means more expensive calculation



Adaptive sampling



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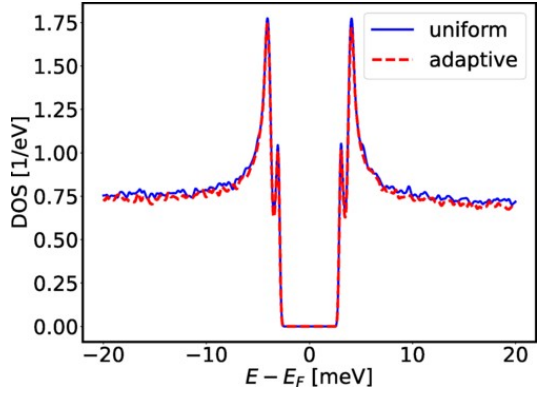
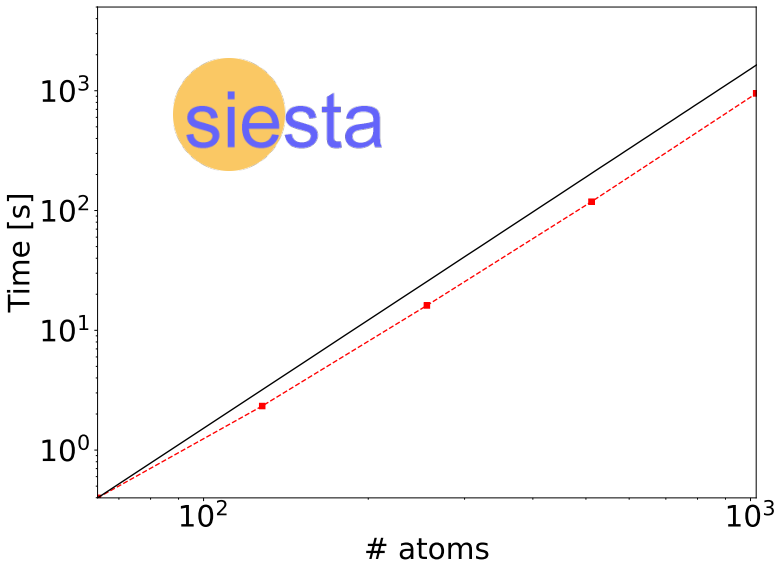


High-performance computing

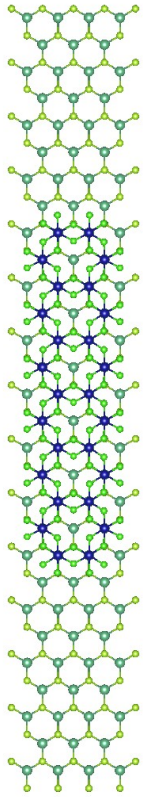
Why we need HPC

Eigenvalue problem

$$\text{time} \propto N_{\text{atom}}^3$$



Heterostructures require large simulations



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Parallellization strategies

Different levels of parallellization

Parallellization strategies

Different levels of parallellization

1) Distributed parallel linear algebra (i.e. ScaLAPACK, ELPA)

3x3 process grid

P1	P2	P3
P4	P5	P6
P7	P8	P9

10x9 matrix

0	1	2	3	4	5	6	7	8
10	11	12	13	14	15	16	17	18
20	21	22	23	24	25	26	27	28
30	31	32	33	34	35	36	37	38
40	41	42	43	44	45	46	47	48
50	51	52	53	54	55	56	57	58
60	61	62	63	64	65	66	67	68
70	71	72	73	74	75	76	77	78
80	81	82	83	84	85	86	87	88
90	91	92	93	94	95	96	97	98

P1

0	1	6	7
10	11	16	17
60	61	66	67
70	71	76	77

P2

2	3	8
12	13	18
62	63	68
72	73	78

P3

4	5
14	15
64	65
74	75

P4

20	21	26	27
30	31	36	37
80	81	86	87
90	91	96	97

P5

22	23	28
32	33	38
82	83	88
92	93	98

P6

24	25
34	35
84	85
94	95

P7

40	41	46	47
50	51	56	57

P8

42	43	48
52	53	58

P9

44	45
54	55

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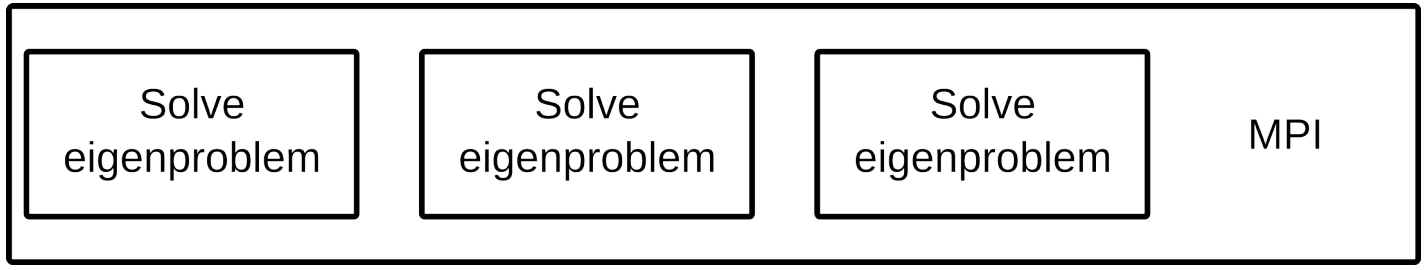
Parallellization strategies

Different levels of parallellization

1) Distributed parallel linear algebra (i.e. ScaLAPACK, ELPA)
2) Solving more eigenvalue problems simultaneously



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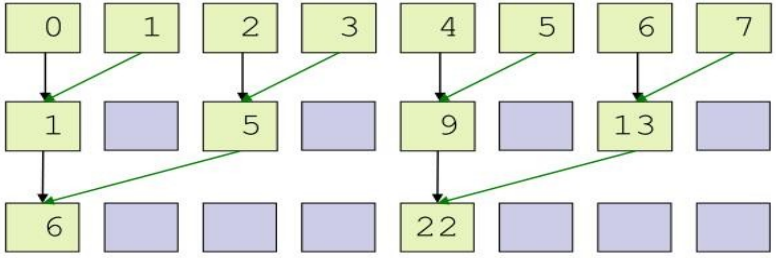
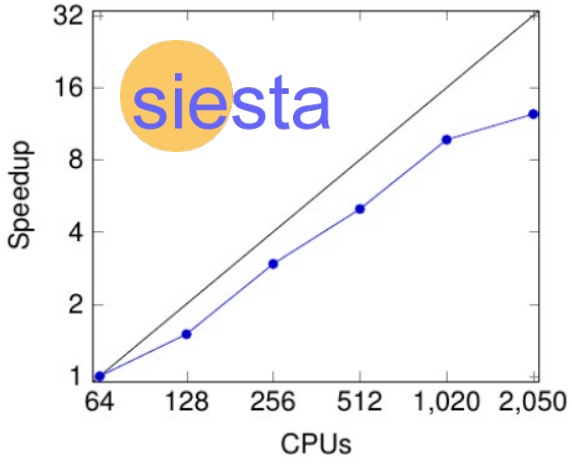
Parallellization strategies

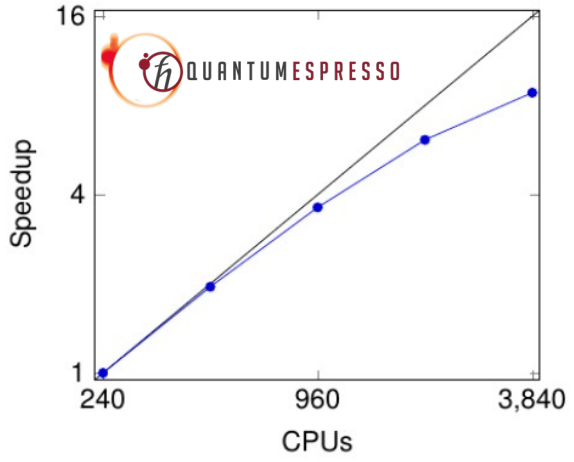
Different levels of parallellization

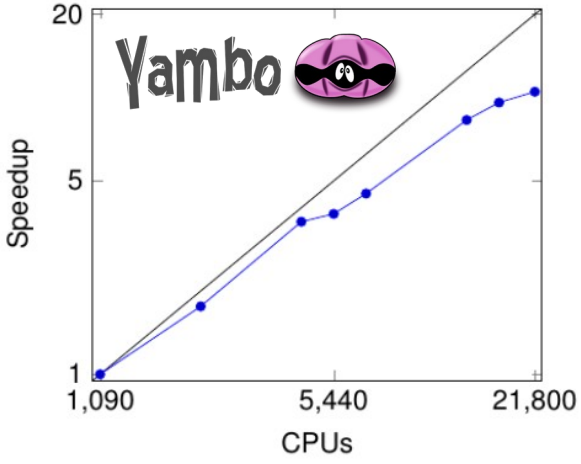
1) Distributed parallel linear algebra (i.e. ScaLAPACK, ELPA)
2) Solving more eigenvalue problems simultaneously
3) Parallel integration

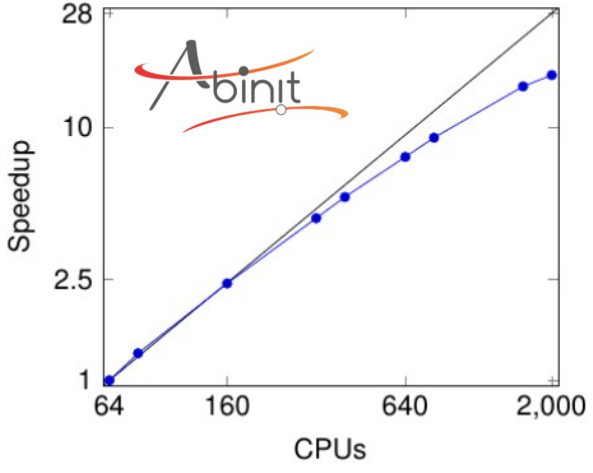


Parallellization strategies





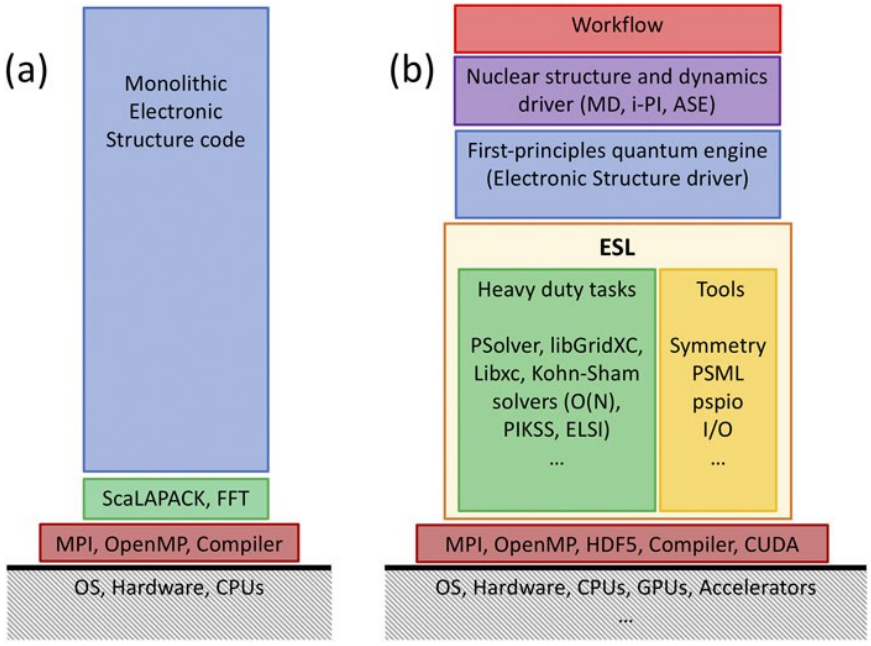




Good practices

Modular code design

Electronic **S**tructure **L**ibrary



J. Chem. Phys.. 2020; 153(2). doi:10.1063/5.0012901

siesta

Abinit

FHI-aims
The ab initio materials simulation package

QUANTUMESPRESSO

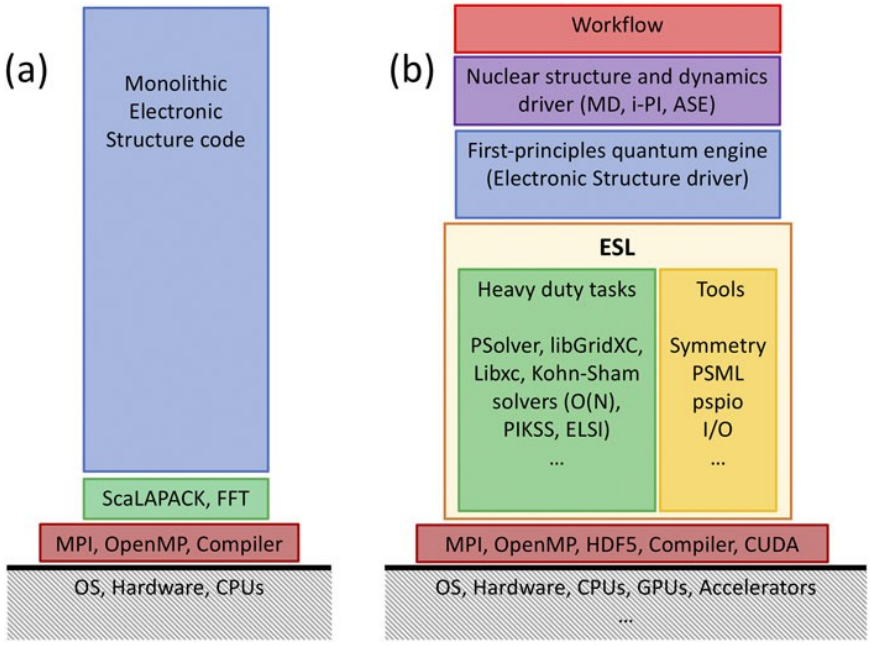
BigDFT

GPAW!

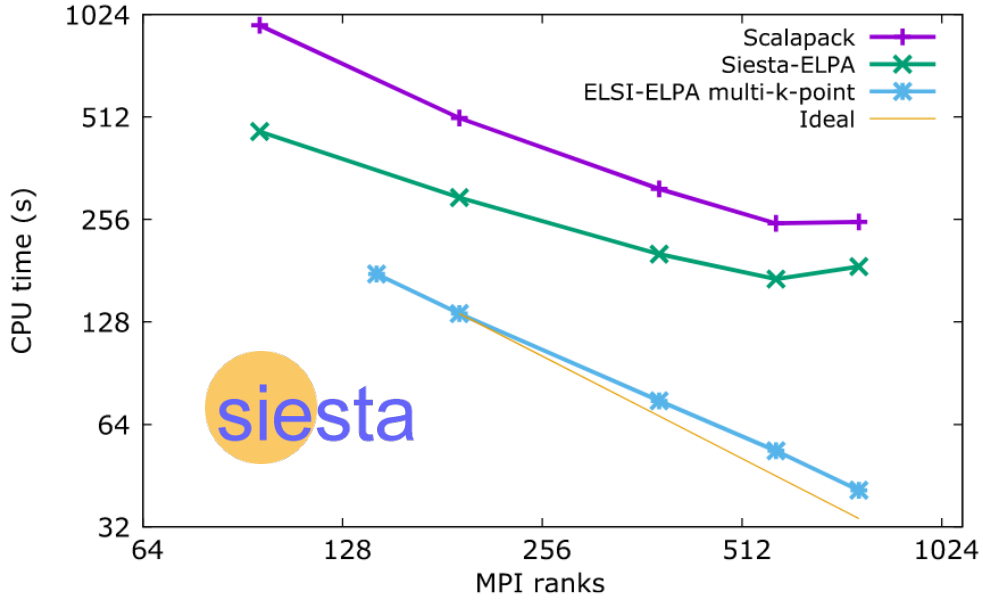
cecam
Centre Européen de Calcul Atomique et Moléculaire

Modular code design

Electronic **S**tructure **L**ibrary



J. Chem. Phys.. 2020; 153(2). doi:10.1063/5.0012901



J. Chem. Phys. 152, 204108 (2020); doi: 10.1063/5.0005077

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ABINIT

FHI-aims
The ab initio materials simulation package

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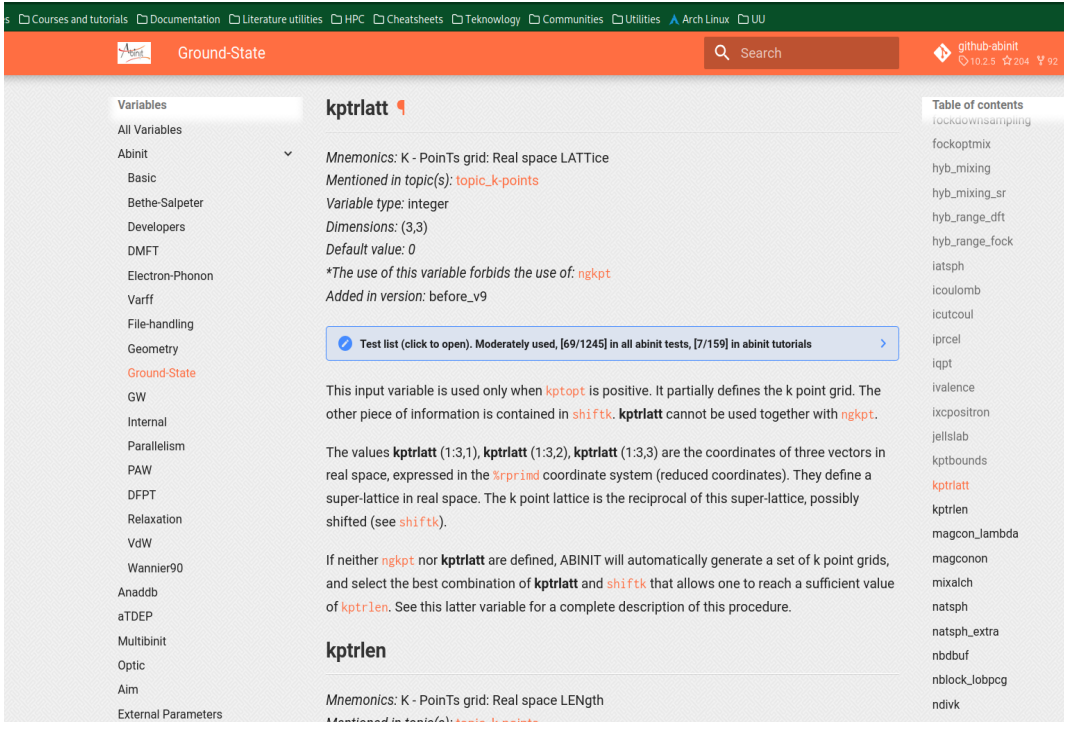
Documentation

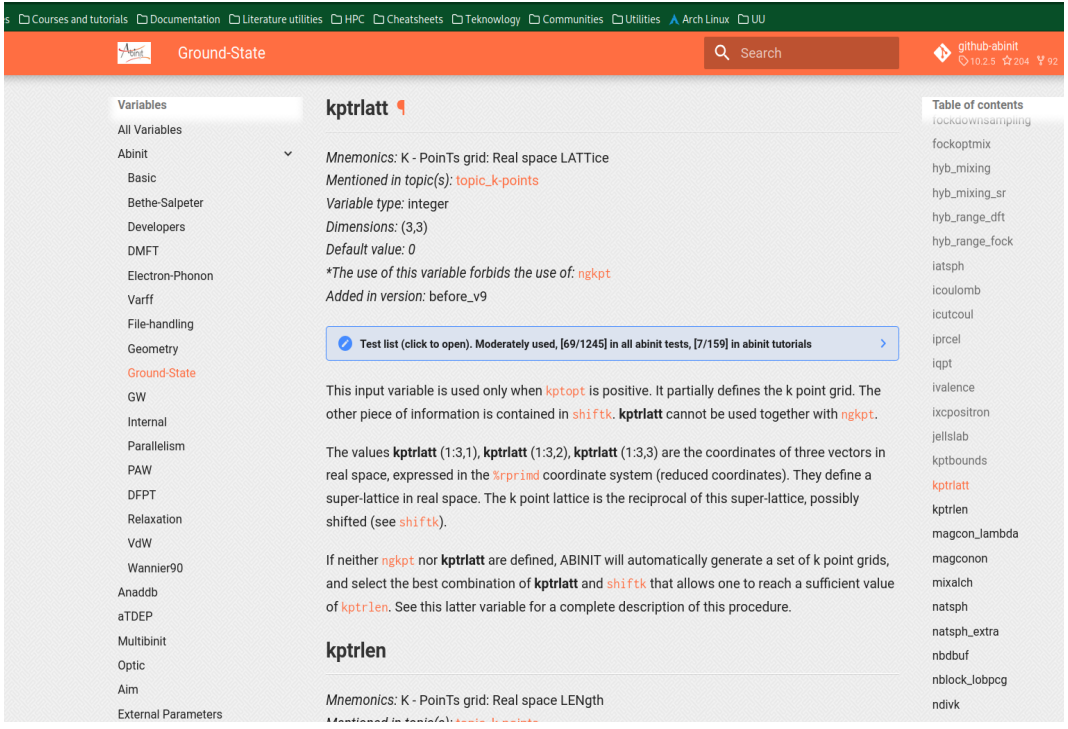
Documentation

Good



Documentation

Good

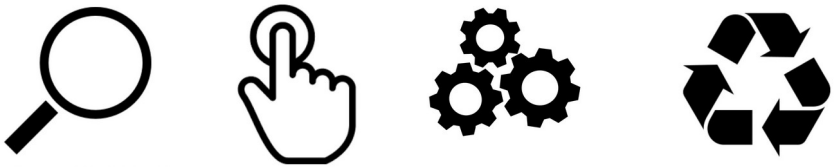


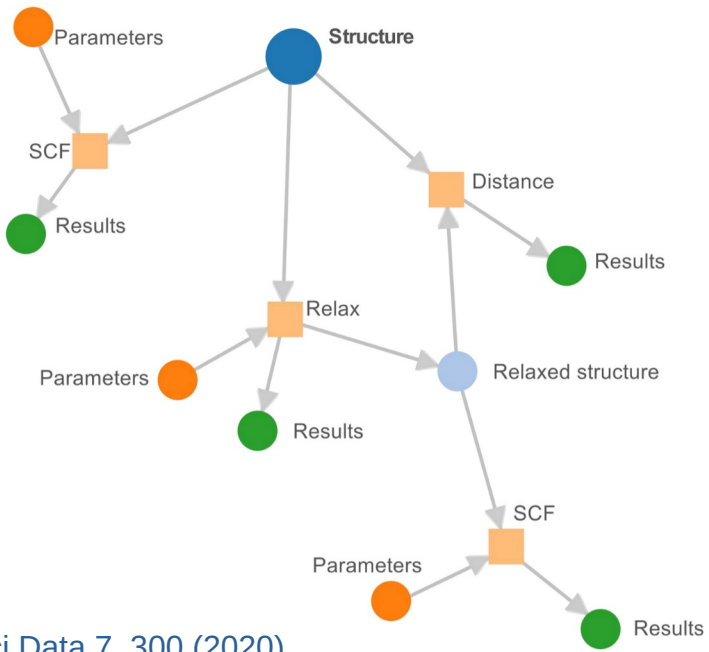
Bad

```
56      stop
57    endif
58
59    call PDPBLASINFO( OUTFILE, nout, M, N, K, nb, NPROW, NPCOL, work, &
60                      iam, NPROCS )
61  !
62  !  Define process grid.
63  !
64    call BLACS_GET( -1, 0, ICTXT )
65    call BLACS_GRIDINIT( ICTXT, 'Row-major', NPROW, NPCOL )
66    call BLACS_GRIDINFO( ICTXT, NPROW, NPCOL, my_row, my_col )
67  !
68  !  Go to bottom of process grid loop if this case doesn't use my
69  !  process.
70  !
71    if ( my_row >= NPROW .or. my_col >= NPCOL ) &
72       go to 20
73
74    MP = numroc( M, nb, my_row, 0, NPROW )
75    KP = numroc( K, nb, my_row, 0, NPROW )
76    KQ = numroc( K, nb, my_col, 0, NPCOL )
77    nq = numroc( N, nb, my_col, 0, NPCOL )
78  !
79  !  Initialize the array descriptor for the matrix A, B and C.
80  !
81    call descinit( desca, M, K, nb, nb, 0, 0, ICTXT, MAX( 1, MP ), &
82                   INFO )
83    call descinit( descb, K, N, nb, nb, 0, 0, ICTXT, MAX( 1, KP ), &
84                   INFO )
85    call descinit( descc, M, N, nb, nb, 0, 0, ICTXT, MAX( 1, MP ), &
86                   INFO )
87  !
88  !  Assign pointers into mem for SCALAPACK arrays, A is
89  !  allocated starting at position mem( 1 ).
90  !
91    IPA = 1
92    IPB = IPA + desca( LLD_ )*KQ
93    IPC = IPB + descb( LLD_ )*NQ
94    IPW = IPC + descc( LLD_ )*NQ
95
96    WORKSIZ = nb
```

FAIR data

Findable Accessible Interoperable Reusable





AiiDA

MATERIALSCLOUD

LEARN WORK DISCOVER EXPLORE ARCHIVE

Open-source Python Infrastructure to
automate, manage, share and reproduce

- Complex computational workflows & data
- Designed for high-throughput workflows
- Interfaced with multiple material science codes
- Ensures full data provenance
- Allows for FAIR data storage

Sci Data 7, 300 (2020)

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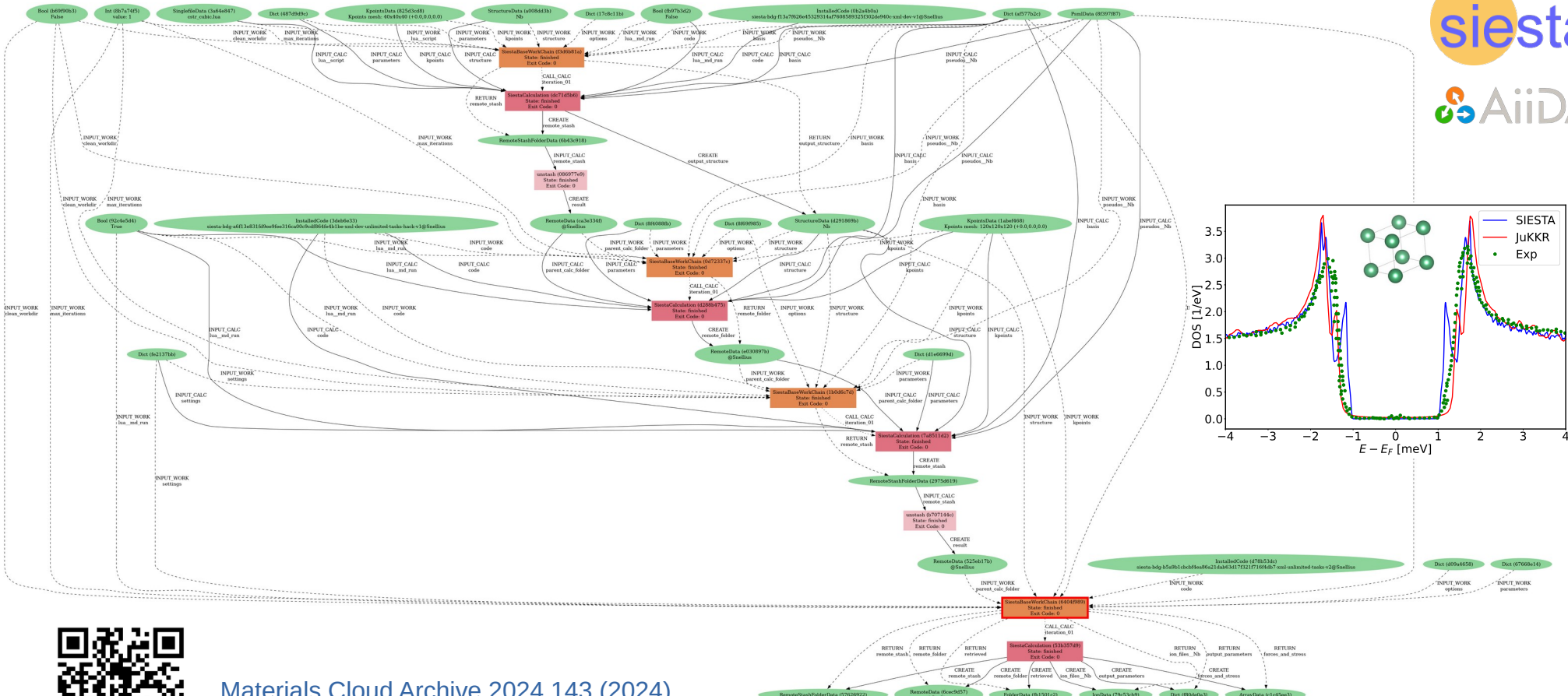
Real world example



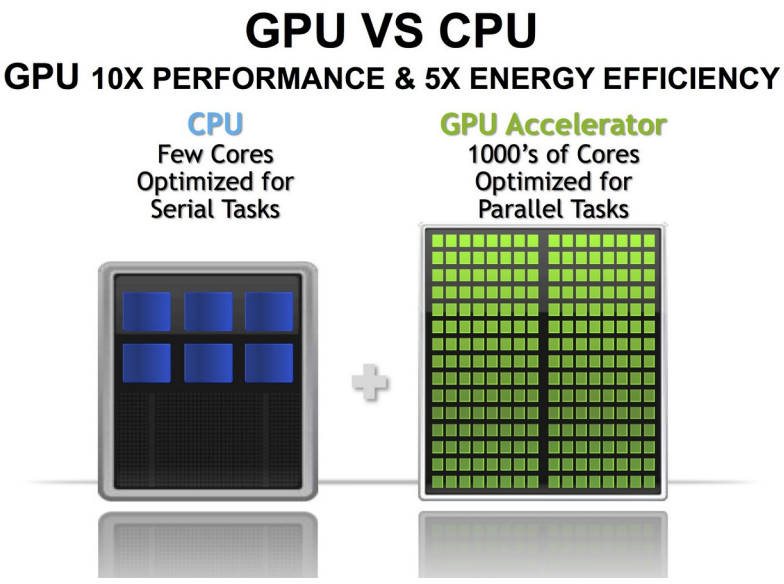
Materials Cloud Archive 2024.143 (2024)

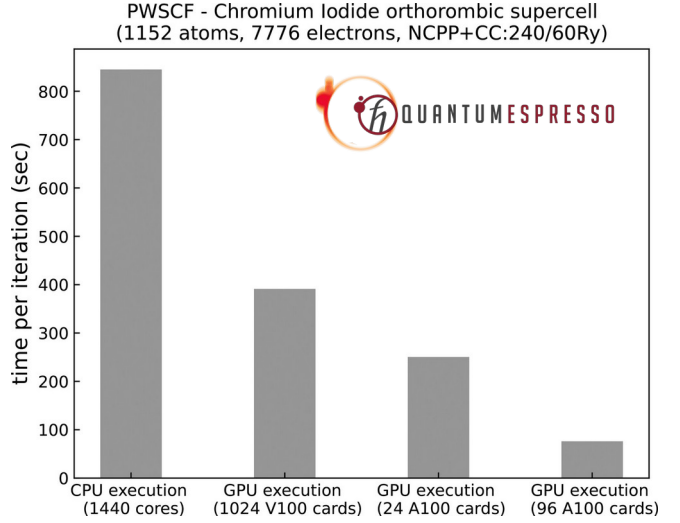
a.h.kole@uu.nl

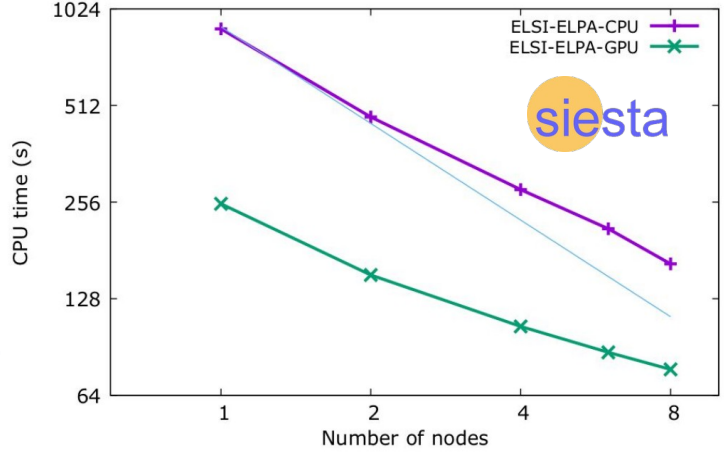
Future

GPU

J. Chem. Theory Comput. 2023, 19, 6992−7006

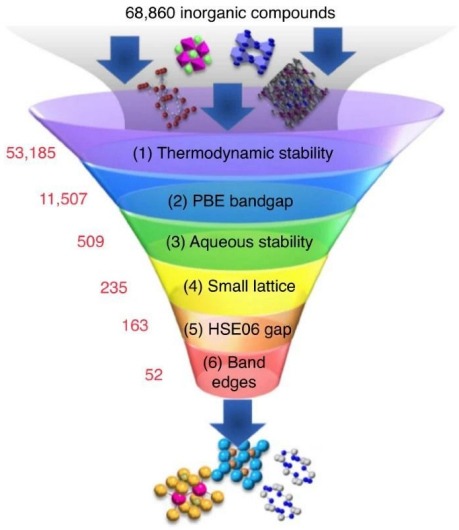


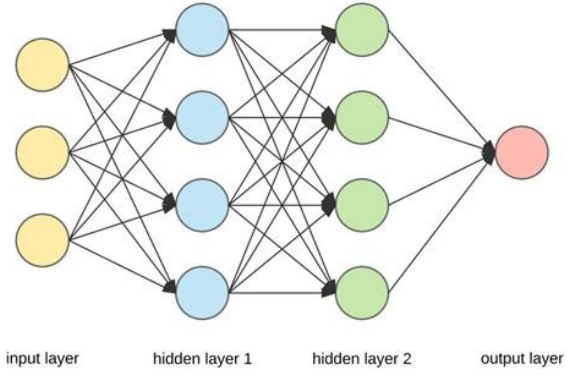


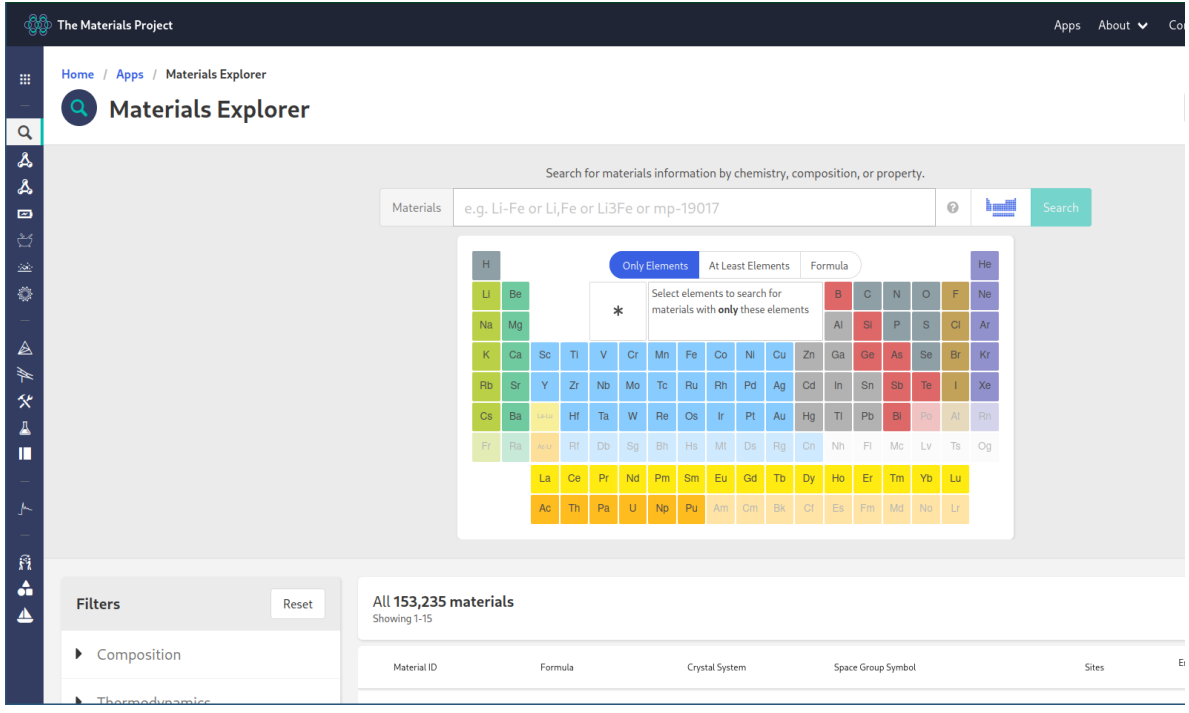


Horizon MaX report 2020

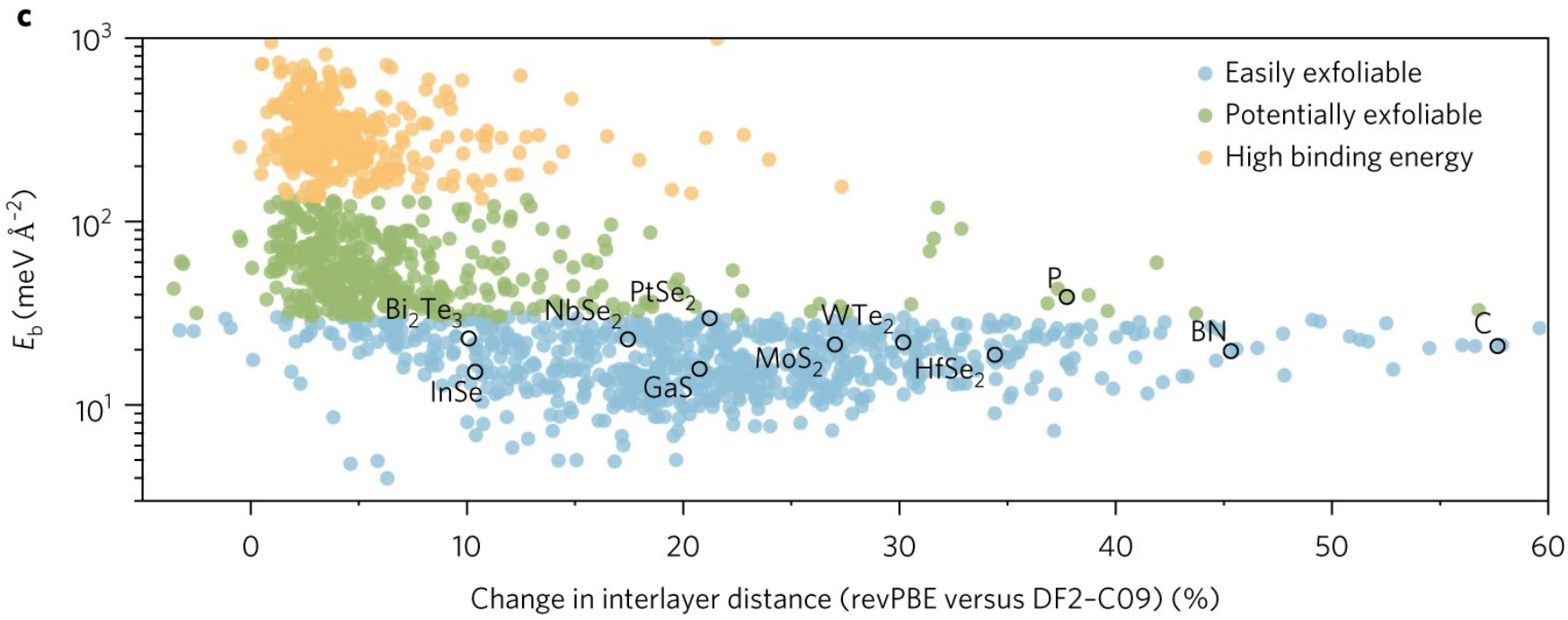
Workflows for high throughput







Workflows for high throughput



Nature Nanotechnology volume 13, pages 246–252 (2018)

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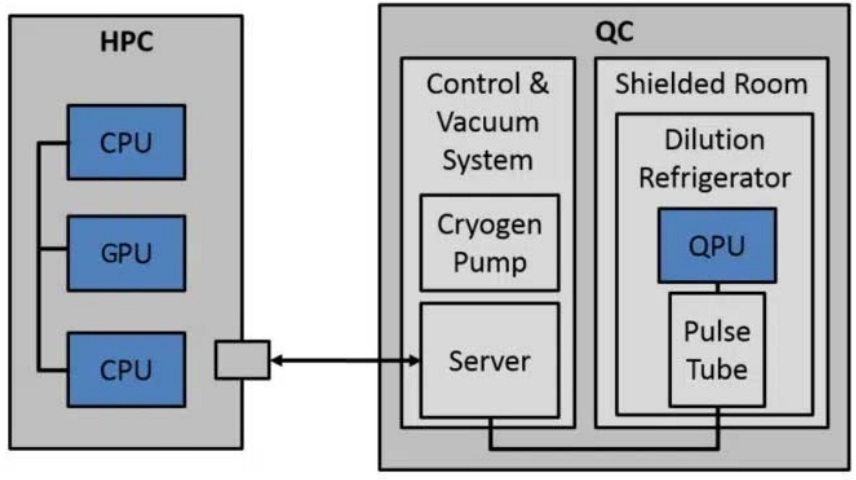
Quantum Computing

- Perfect fit for solving quantum problem
- Current applicability still limited => hybrid algorithms
- Using regular (CPU+GPU) and quantum (QPU) hardware in tandem



Summary

- Quantum Materials exhibit quantum behaviour at macroscopic scales
- Quantum description using first-principles simulations
- Compute properties in a parameter-free way to aid materials design
- Recent extension to include superconductivity
- Open-source codes that scale well on High Performance Computing infrastructure
- Good practices to help the community

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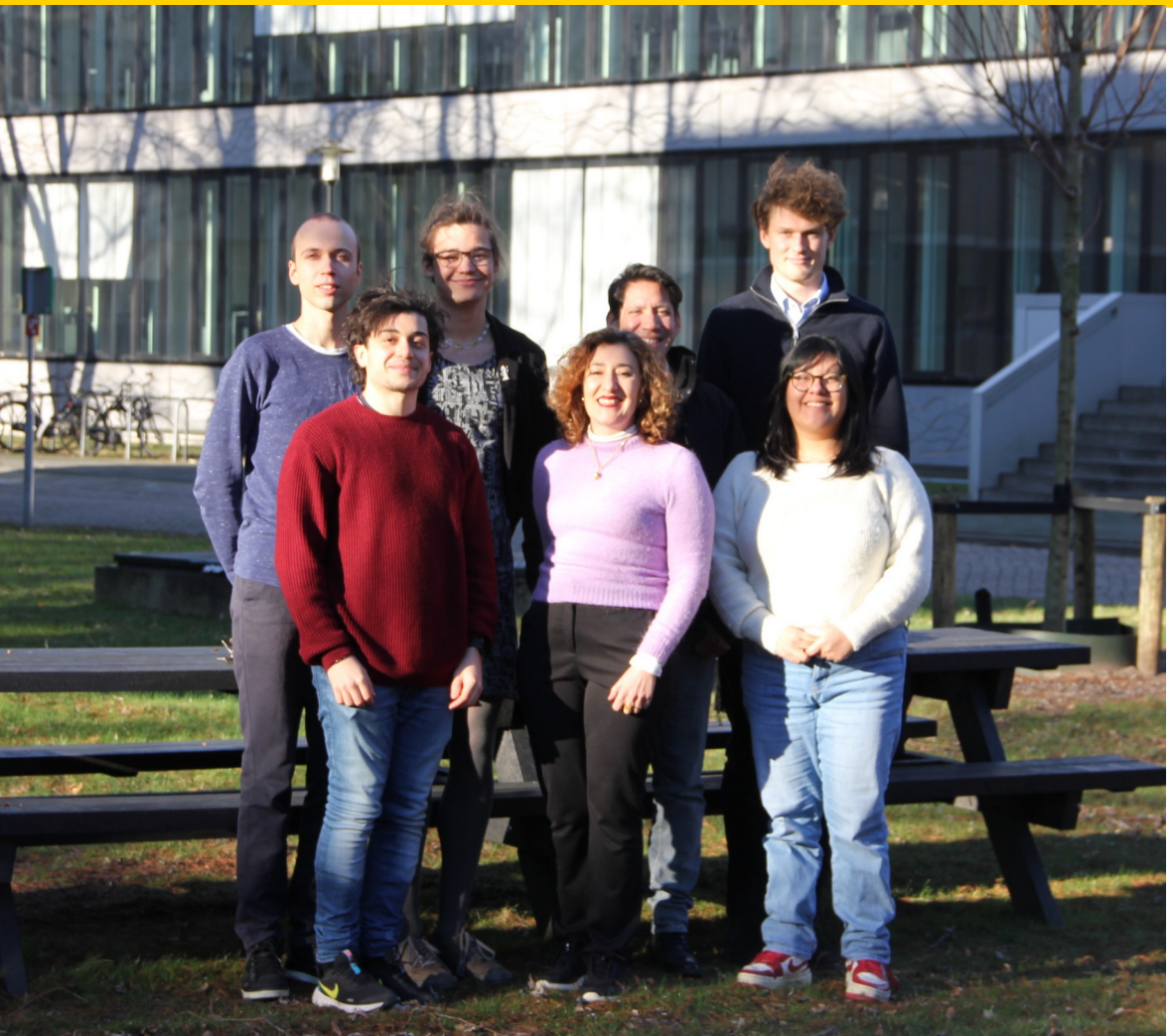
Quantum Materials by Design 2024 Team



Funding

QuMat
Materials for the Quantum Age

NWO

SURF

QUONDENSATE

European Innovation Council

PRACE
PARTNERSHIP FOR ADVANCED COMPUTING IN EUROPE

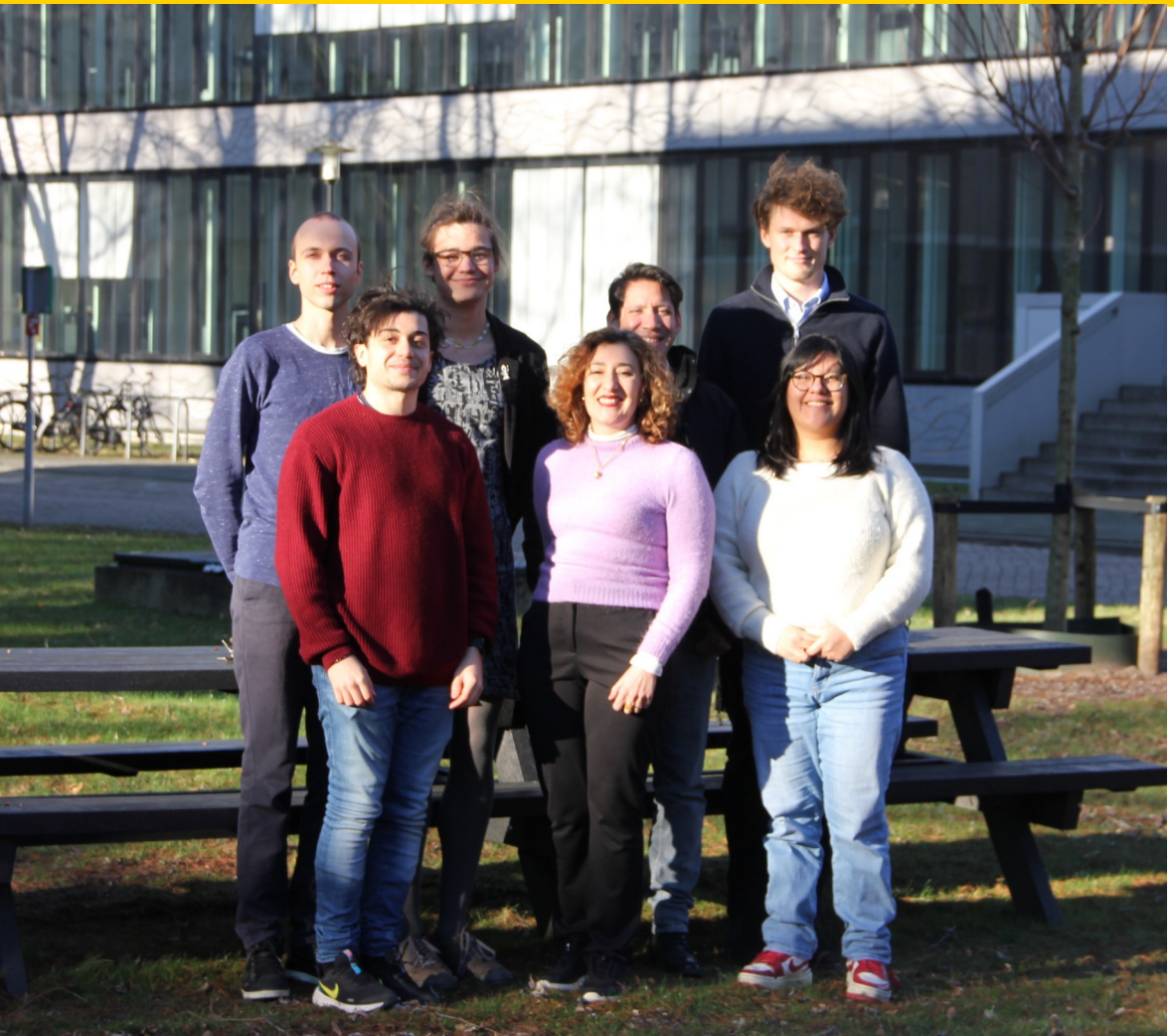
Codes

siesta

QUANTUMESPRESSO

Yambo

Quantum Materials by Design 2024 Team



Projects on Snellius

- NWO-2022.012
- EINF-6220
- NWO-2024.012

Publications

Sohier 2D Mat 10(2), 025006, 2023
Melo npj CM 9(1), 147, 2023
Zanolli A&A 675, L9, 2023
Moes NL 24(17), 5110-5116, 2024
Reho PRB 110 (3), 035118, 2024
Reho PRB 110 (13), 134505, 2024
Reho arxiv 2024
Cuperus submitted 2024
Vonk submitted 2024
Reho in preparation 2025

Student theses

Wittemeier 2023
Tops 2024
Dubbink 2024

14 keynote and invited presentations at international conferences

Questions?