MANY-BODY POST-PROCESSING OF DFT CALCULATIONS USING VQE

Erik Schultheis, Alexander Rehn, Gabriel Breuil

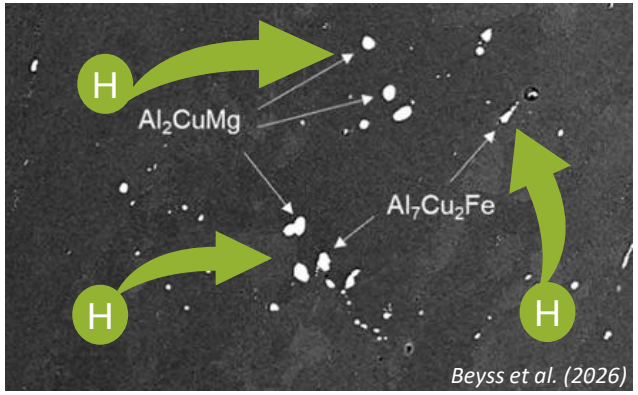
German Aerospace Center (DLR), Institute of Frontier Materials on Earth and in Space, Cologne

From Quantum Mechanics to Material Science and Engineering

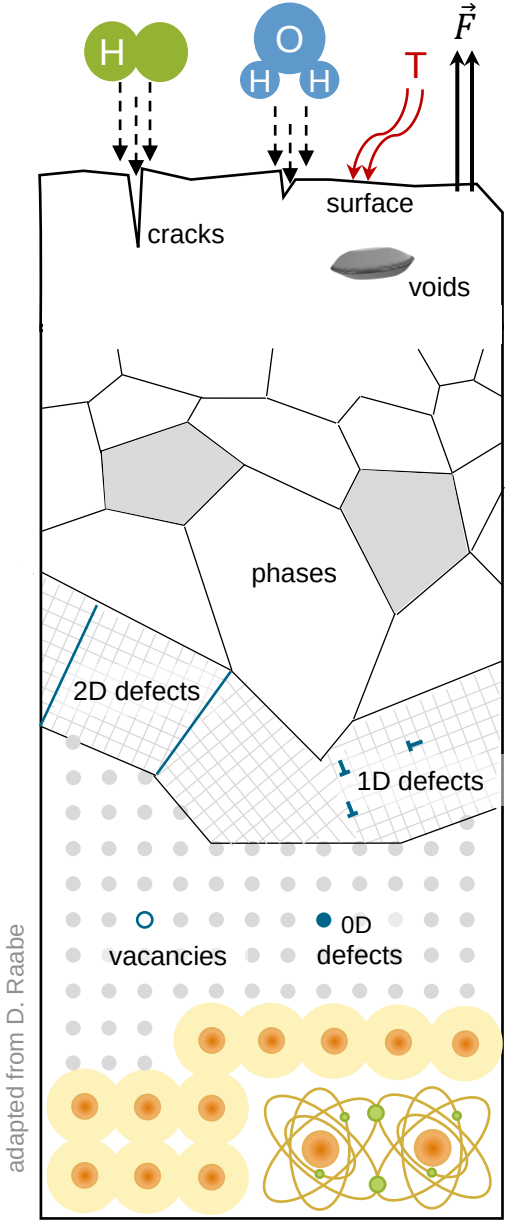
Applications



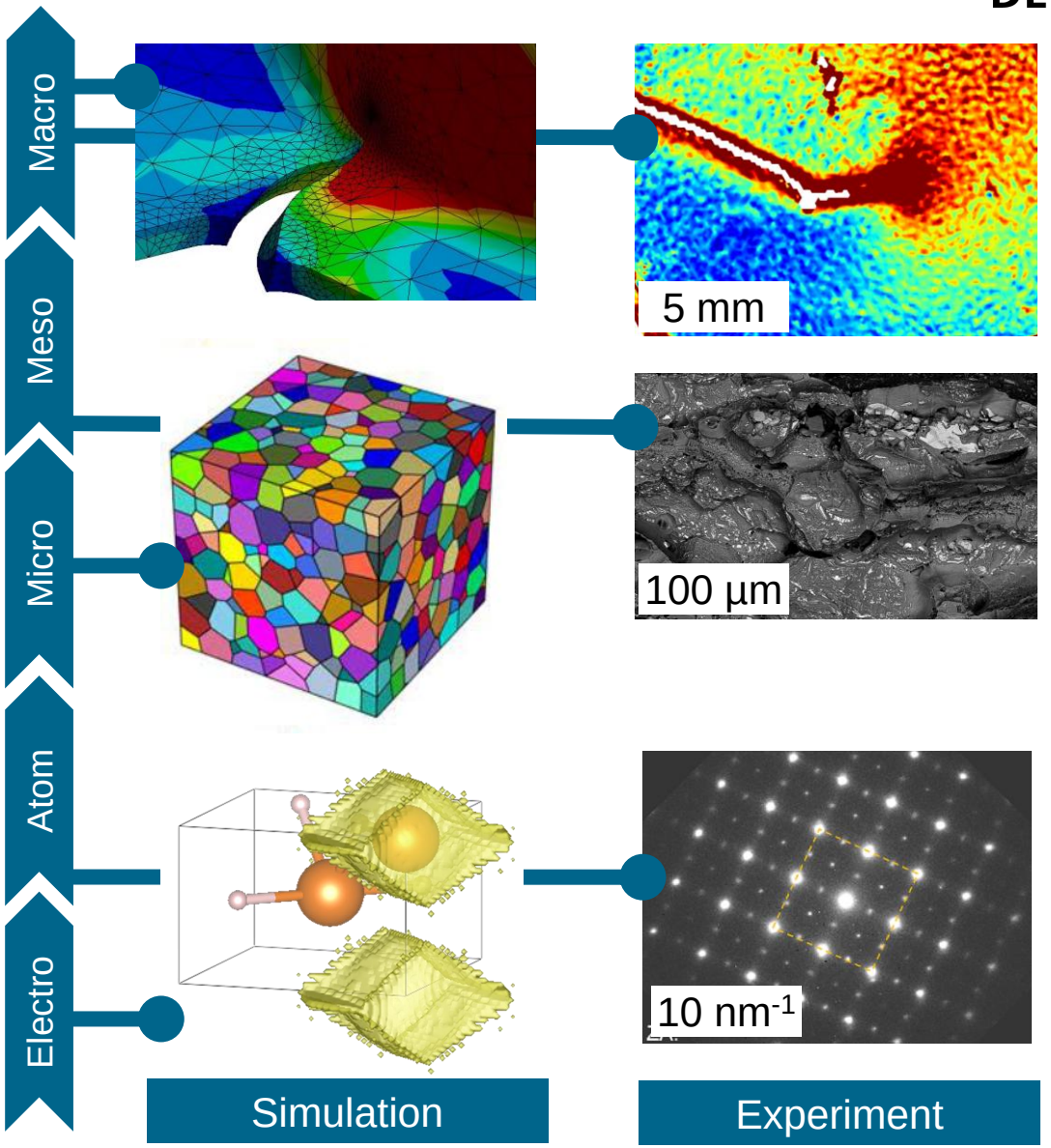
Corrosion



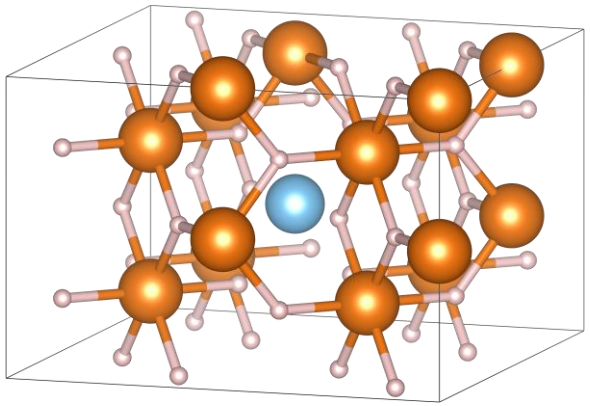
H-Embrittlement



adapted from D. Raabe

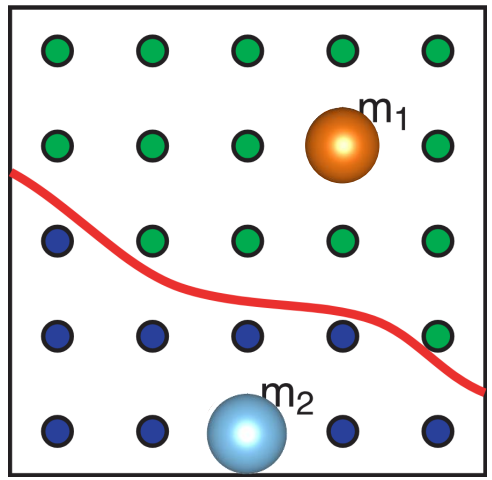


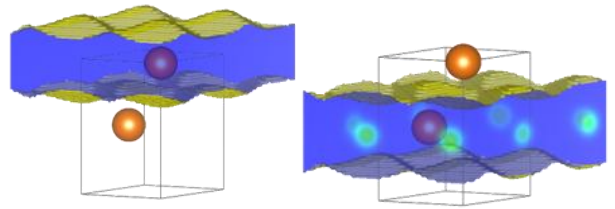
Material Characterization via Bader Charges



MgH_2-TM

$$Q_I = \int_{B_I} \rho(\mathbf{r})\, \mathrm{d}^3\mathbf{r}$$

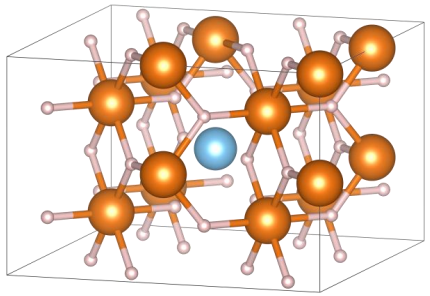


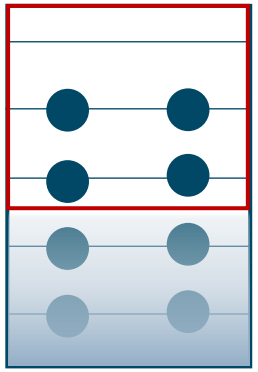


Bader charges quantify:

- Density partitioning among atoms
- Charge redistribution
- Bonding
- Oxidation states
- Charge locality and charge transfer

Quantum-Ready Computational Workflow





$$\hat{H}^{(k)}_{\text{elec}} = \sum_{tu} h^{(k)}_{tu} \hat{a}^\dagger_t \hat{a}_u + \frac{1}{2} \sum_{tuvw} h^{(k)}_{tuvw} \hat{a}^\dagger_t \hat{a}^\dagger_u \hat{a}_v \hat{a}_w + E_{\text{n-n}} + E_{\text{e-self}}$$

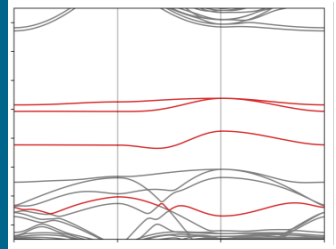


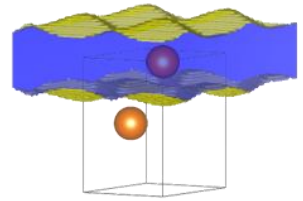
DFT

Selected Kohn-Sham orbitals

Active-Space Hamiltonian

CASCI VQE





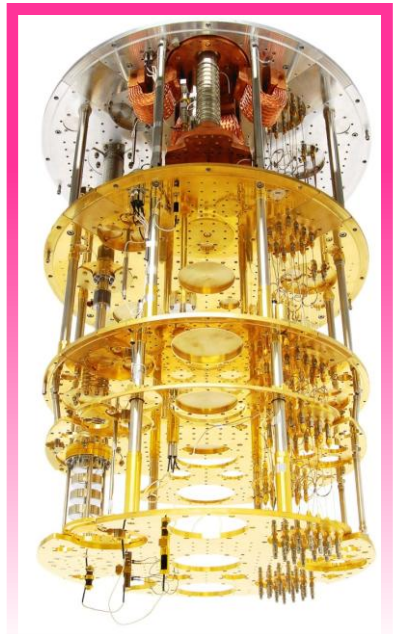
Electron density

$\rho(\mathbf{r})$

Bader charges

Erik Schultheis, DLR Institute of Frontier Materials on Earth and in Space, 30.09.2025

VQE: Finding the ground state



$|\psi(\theta)\rangle, H$

$\langle\psi(\theta)|H|\psi(\theta)\rangle$

Classical Optimizer

$$\theta^* = \arg\min_{\theta} \langle\psi(\theta)|H|\psi(\theta)\rangle$$

Converged?

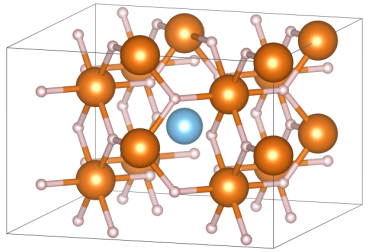
Optimized state → Electron density

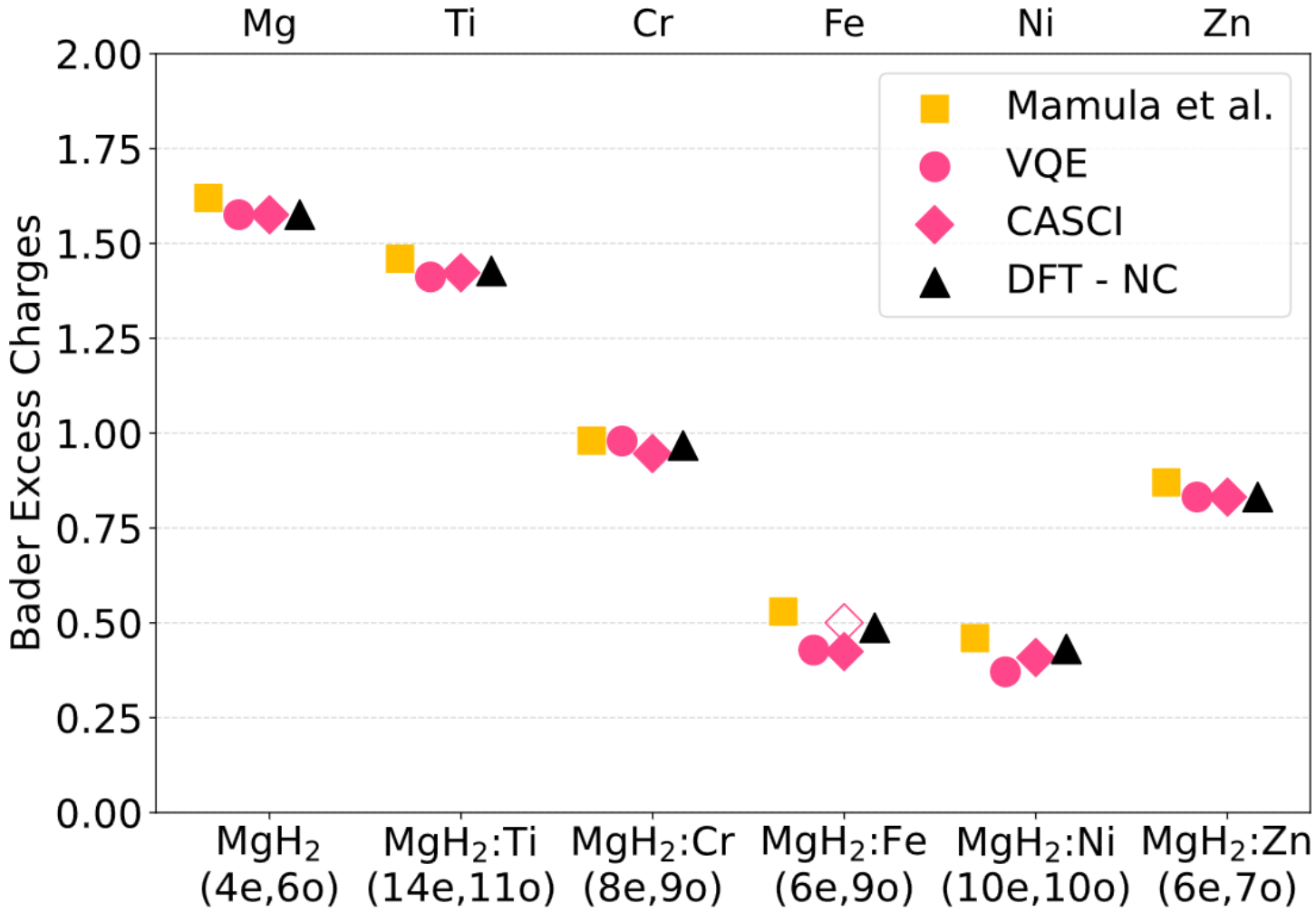


adapted from
Chen et al., arXiv:2405.11687

VQE simulated classically in this work. No quantum hardware used

Bader charges: MgH_2-TM

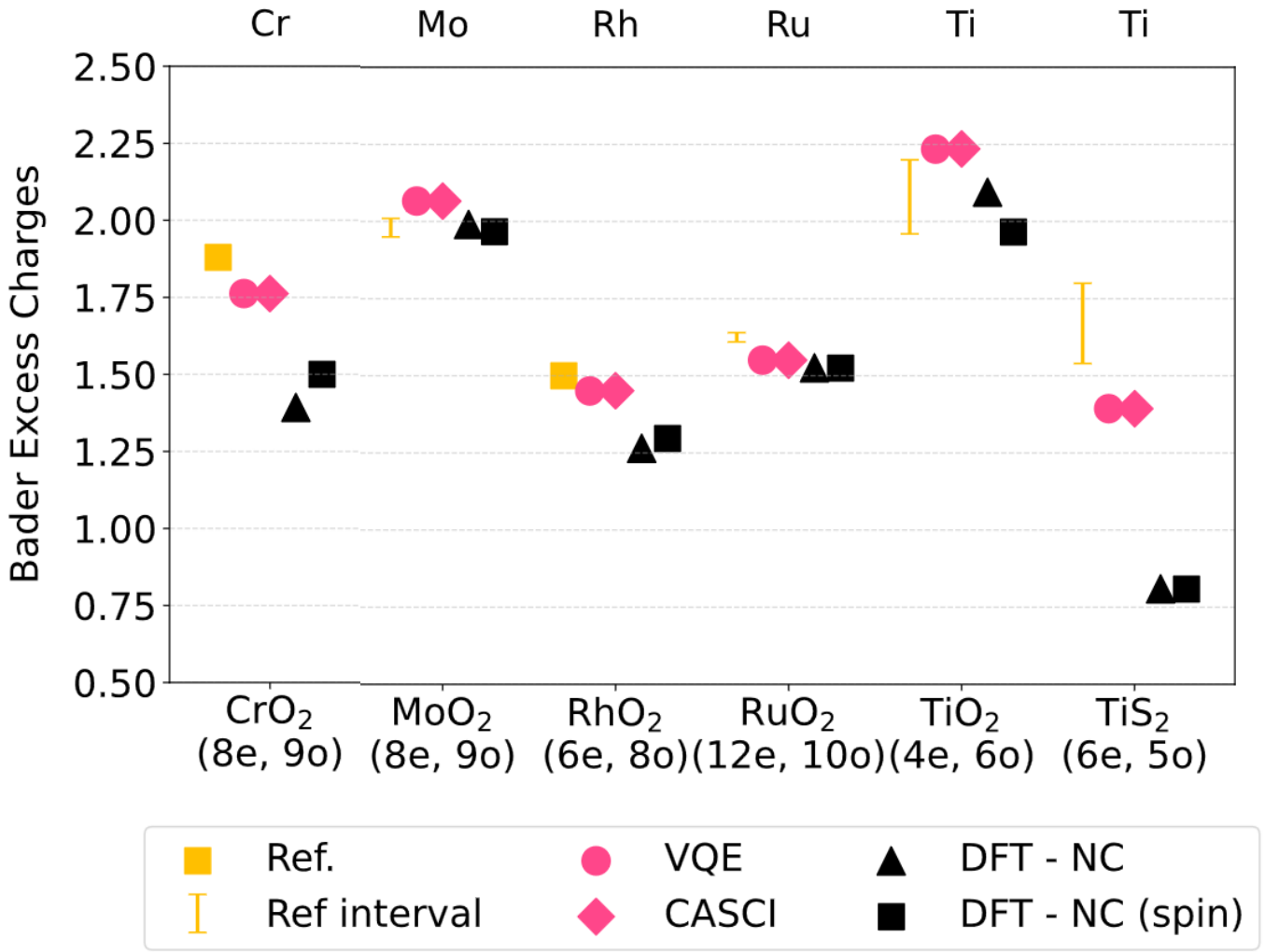




Bader charges match DFT and DFT literature values

Schultheis et al., arXiv:2510.12887

Bader charges: Transition metal compounds



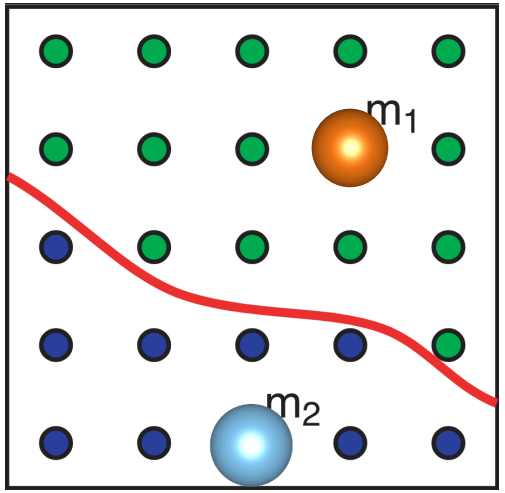
Bader charges material-dependent. Improvements towards reference DFT+U values visible

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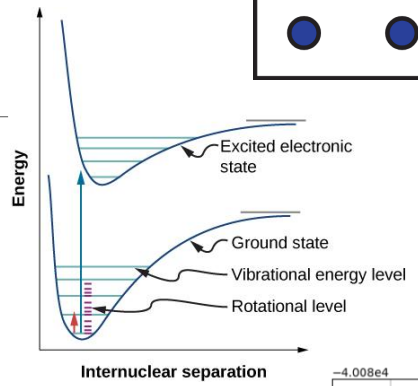
Schultheis et al., arXiv:2510.12887

Summary and future work

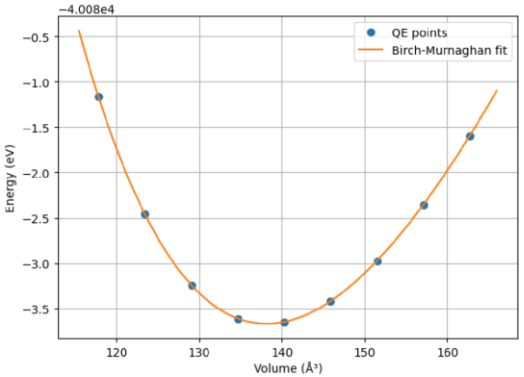
- Improvement of Bader charges towards selected DFT+U references for strongly correlated materials
- Software available: Dopyqo
  - Generates and solves Hamiltonians of periodic materials



- Bader charge dependence on active space



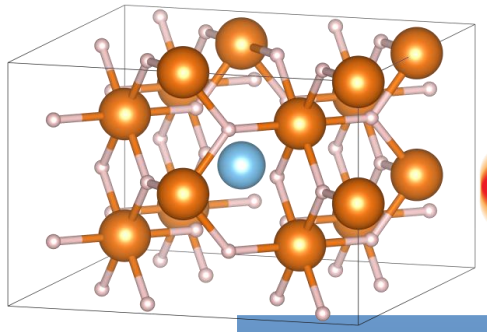
- Investigating other properties: bulk modulus, excited states



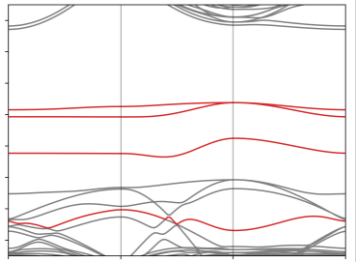
- Characterizing hydrogen embrittlement in aerospace materials

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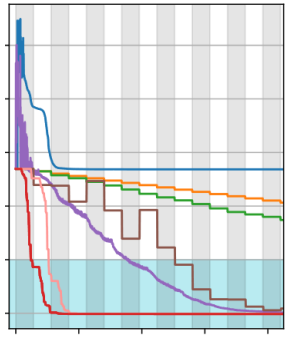
Questions?

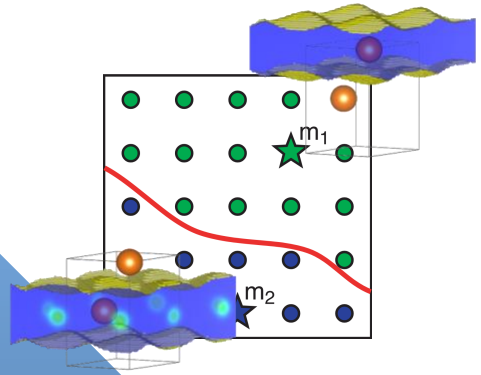






$$\hat{H}^{(\mathbf{k})}_{\text{elec}} = \sum_{tu} h^{(\mathbf{k})}_{tu} \hat{a}^\dagger_t \hat{a}_u + \frac{1}{2} \sum_{tuvw} h^{(\mathbf{k})}_{tuvw} \hat{a}^\dagger_t \hat{a}^\dagger_u \hat{a}_v \hat{a}_w + E_{\text{n-n}} + E_{\text{e-self}}$$





Material → DFT Simulation → Many-body Hamiltonian → Ground state → Properties

Dopyqo

pip install dopyqo

DLR-WF/Dopyqo

Erik Schultheis, DLR Institute of Frontier Materials on Earth and in Space, 30.09.2025

Schultheis et al., arXiv:2510.12887

Calculating charges on atoms: Bader charges

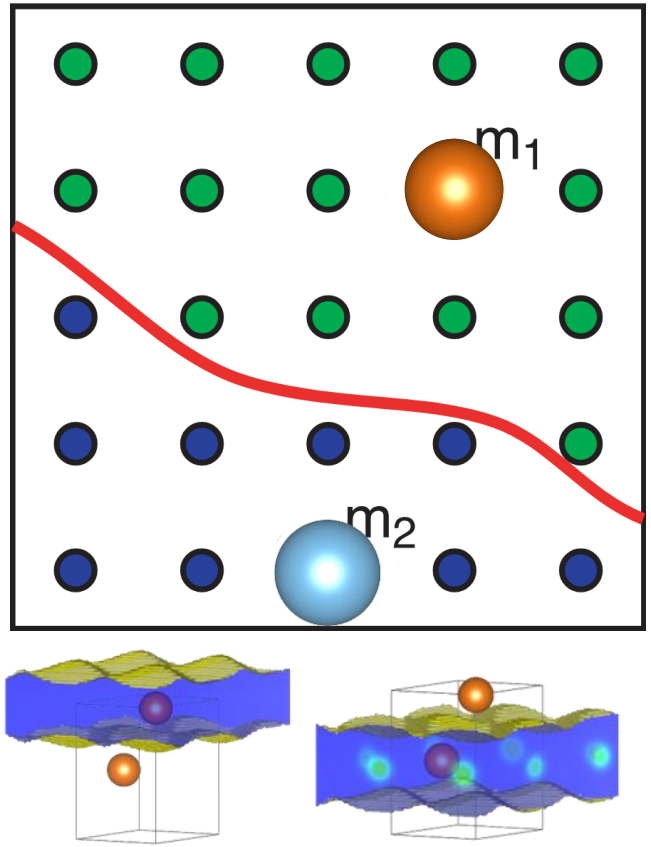
- Bader regions
  - Real-space is divided into regions
    bound by zero-flux surfaces
    running through minima of the charge density
- Bader (excess) charge:

$$Q_I = \int_{B_I} \rho(\mathbf{r})\,\mathrm{d}^3\mathbf{r} \qquad Q_I \to Q_I - Z_I$$

- From density:

$$\rho(\mathbf{r}) = \sum_{tu} \gamma_{tu} \psi_t^*(\mathbf{r}) \psi_u(\mathbf{r})$$

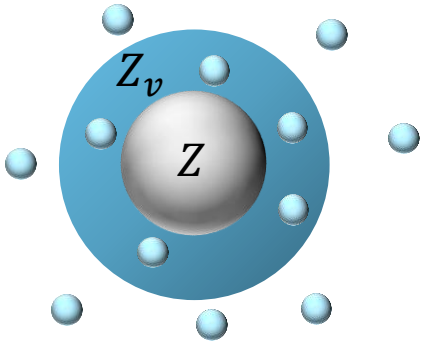
Diagonal in DFT: $\rho(\mathbf{r}) = \sum_t f_t \left|\psi_t(\mathbf{r})\right|^2$

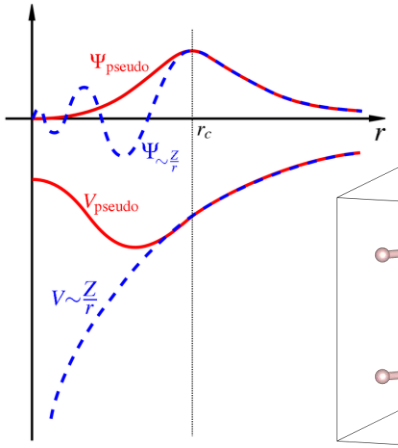


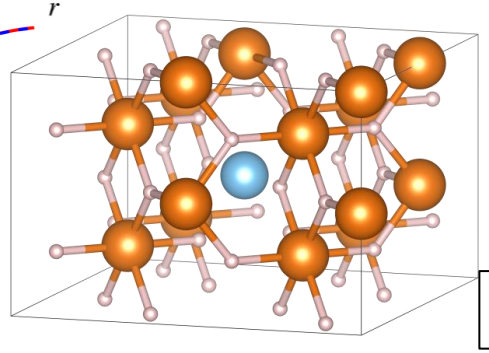
Simulation of periodic systems: Many-body approach

$$H = -\frac{1}{2}\sum_{T,i}\nabla^2_{r_i+T} + \sum_{T,I,i} V_{\mathrm{pp}}(r_i - R_I - T, r_i' - R_I - T) + \sum_{T,I,J}\frac{Z_I Z_J}{|R_I + T - R_J|} + \sum_{T,i,j}\frac{1}{|r_i + T - r_j|}$$

$$\hat{H} = \text{kin. energy} + \text{pseudopot.} + \text{nuclear int.} + \text{electron int.}$$







Kinetic energy of electrons (Born-Oppenheimer Approximation)

Potential of nuclei + core-electrons

Fully non-local norm-conserving **pseudopotential**

Energy between all nuclei and their (infinitely many) periodic images

Calculated via Ewald summation

Exact coulomb interaction between electrons

Calculated via pair densities

All calculated with KS-orbitals in plane-wave basis

$$|\psi_i\rangle = \sum_p c_{p,i}\,|p\rangle$$

plane-wave

DFT Hamiltonian is discarded

Periodicity through sum over lattice translation vectors T

Schultheis et al., arXiv:2510.12887