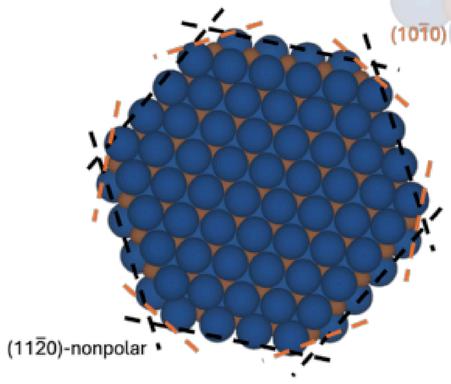
Towards more realistic atomistic models of MXene flakes



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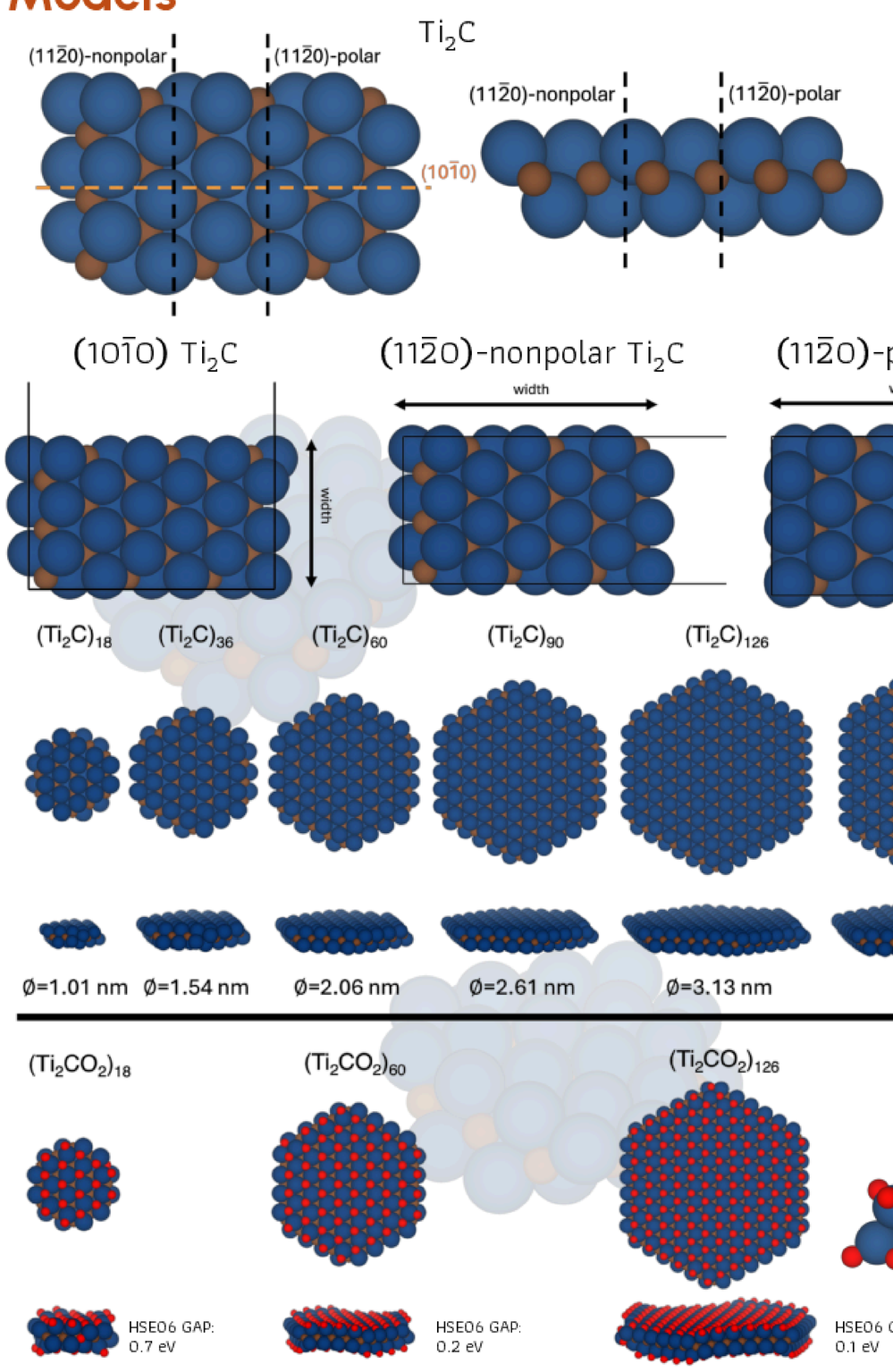
Motivation

MXenes are 2D materials, $M_{y+1}X_yT_x$, where M = early transition metal, X = C and/or N, y = 1–4, and T_x = none, F, Cl, H, O, and OH, are chemical groups functionalizing the surface.[1]

They exhibit diverse **properties** and **applications**, tunable through composition, thickness, and functionalization, such as a thermo-, electro-, and photo-catalysis.[2,3]

Provide a **more realistic representation** using the **Wulff** construction,[4] to model **finite $(Ti_2C)_n$ MXene flake** models, where n = number of Ti_2C units.

Models



Computacional Details

- Geometrical relaxations with **PBE**.
- Numerical Atomic-centred Orbitals with **tier-1/light-grid + scalar relativistic effects.**
- Smearing of 0.01 eV Gaussian broadening.
- Electronic threshold : 10^{-5} eV.
- Periodic calculations at 7×7×1 **k**-points + 30Å of vacuum.
- Converged optimizations when the forces acting on the nuclei are <0.001 eV·Å^{-1}.
- Single points with **HSE06**.

FHI-aims The ab initio materials simulation package

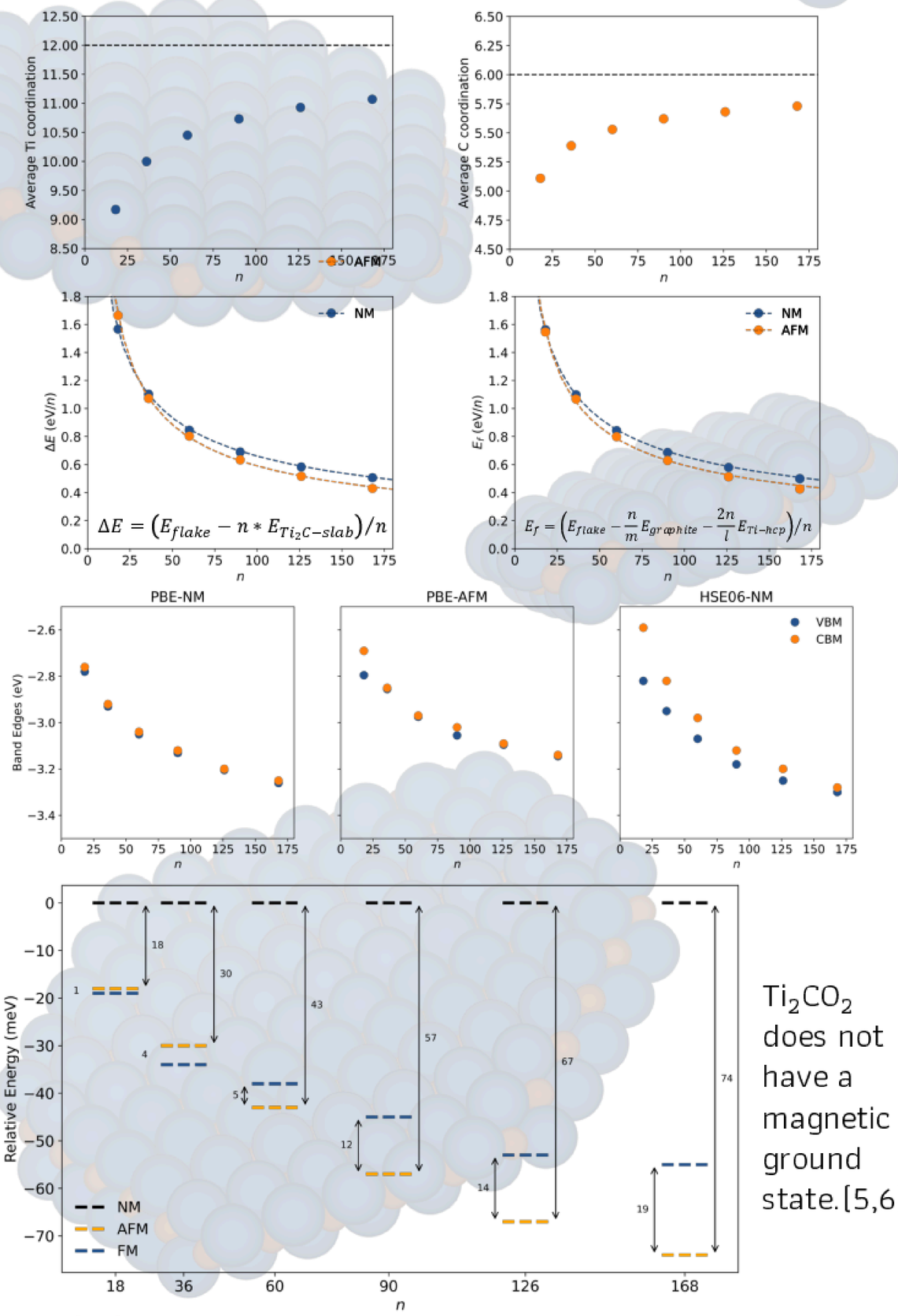
J. M. Rahm & P. Erhart, *J. Open Source Softw.*, **2020**, *5*, 1944.

Surface Energies

$$\gamma = \frac{E_{nanorib} - n * E_{Ti_2C-slab}}{2S}$$

Surface	γ [J/m²]
(11$\bar{2}$0)-nonpolar	4.63
(11$\bar{2}$0)-polar	5.79
[10$\bar{1}$0]	4.93

Results



Ti_2CO_2 does not have a magnetic ground state.[5,6]

Conclusions

- Ti_2C flakes show **hexagonal** shapes and converge in structure and energy to periodic slabs, with **AFM** as the stable ground state for larger flakes.
- All flakes remain **gapless** with negligible quantum confinement (QC) effects.
- O-functionalized flakes remain non-magnetic, slightly **deformed**, and show **size-dependent bandgap** narrowing related to edge effects.
- **QC** significantly affects **small flakes**' properties but fades with size.

References

[1] Y. Gogotsi, B. Anasori, *ACS Nano.*, **2019**, *13*, 8491–8494.
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[3] R. Ramírez Grau, *et al.*, *J. Am. Chem. Soc.*, **2025**, *147*, 3315–3332.
[4] G. Wulff, *Z. Kristallogr.*, **1901**, *34*, 449–530.
[5] N. García-Romeral, *J. Phys. Chem. C*, **2023**, *127*, 3706–3714.
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Aknowledgements

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