Dynamic Charge Transfer Potential for Modeling Oxide Interfaces

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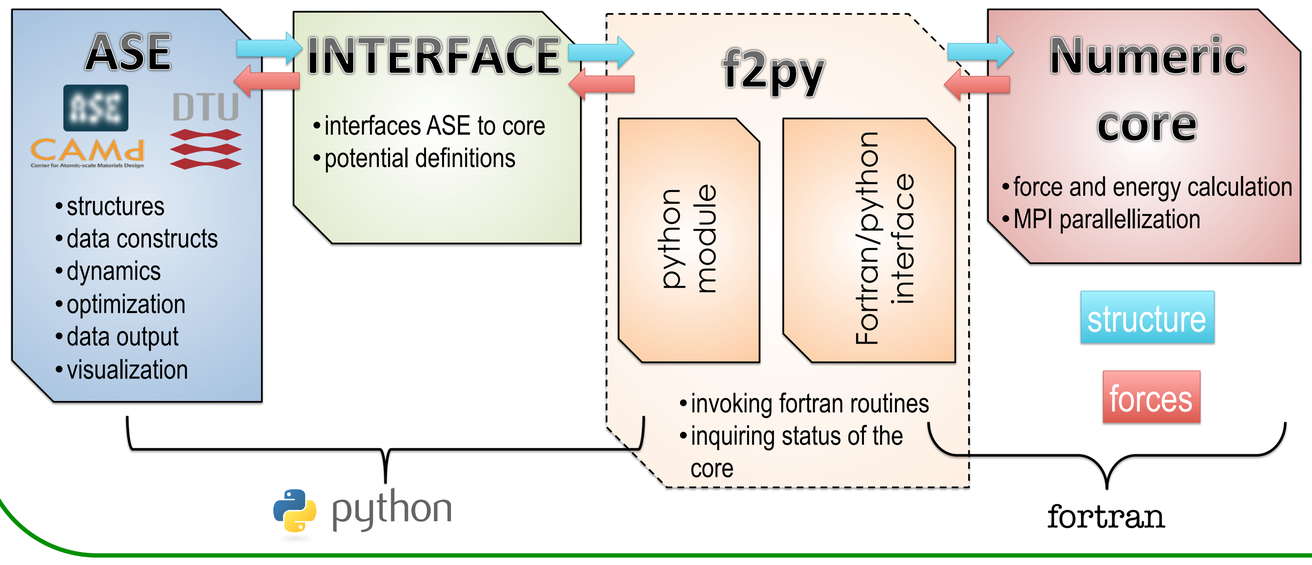
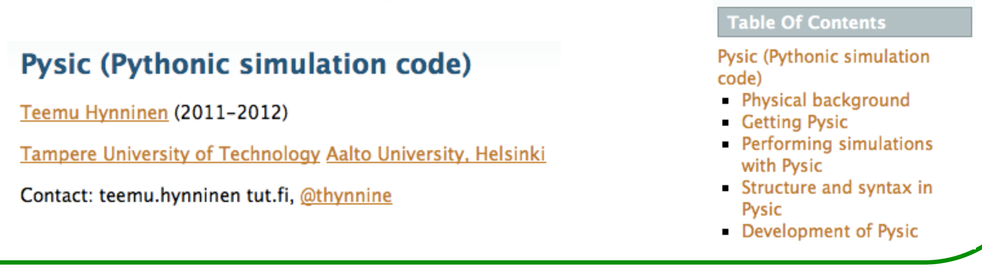
Pysic

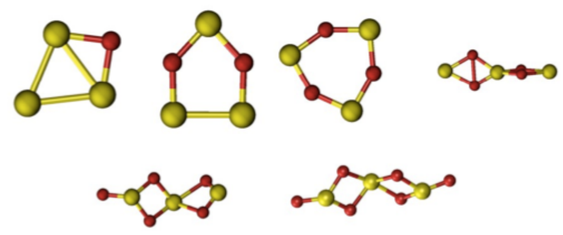


- Pysic as a module of Python (library of tools)
- Advanced variable charge potentials implementation (equilibration of atomic charges)
- Used for MD simulations of interfaces using different bond order potentials
- Pysic 0.4.4. documentation available at: http://thynnine.github.com/pysic/
- Source code available at: https://github.com/thynnine/pysic/



Si_3O_y (y = 1:6) Clusters Analysis

- Clusters allow to obtain information related to different charge states by modifying both the number and stoichiometric ratio of the atoms contained within
- Focus on these systems because of the importance of the Si/SiO_2 interface



COMB Potential

- Empirical Potential with ~20 parameters
- Variable Atomic Charges
- Many-body Interactions
- Parameterized for Si, O and Hf
- Implemented in Pysic (with COMB10 parameters)

$$E_i^S(q_i) = \chi_i q_i + J_i q_i^2 + K_i q_i^3 + L_i q_i^4$$

$$E_{\text{Si-O-Si}} = \sum_i \sum_{j\neq i} \sum_{k\neq i,j} f_{C_{ij}} f_{C_{ik}} K_{\text{Si-O-Si}} (\cos\theta_{\text{Si-O-Si}} - \cos\theta^0_{\text{Si-O-Si}})^2$$

Self-Energy — Bond-Bending

$$E_T = \sum_i \left[E_i^S + \frac{1}{2} \sum_{j\neq i} V_{ij}(r_{ij}, q_i, q_j) + E_i^{BB} \right]$$

Potential

$$V_{ij}(r_{ij}, q_i, q_j) = U_{ij}^R(r_{ij}) + U_{ij}^A(r_{ij}, q_i, q_j) + U_{ij}^I(r_{ij}, q_i, q_j),$$

$$U_{ij}^R(r_{ij}) = f_{S_{ij}} A_{ij} e^{(-\lambda_{ij} r_{ij})},$$ Repulsive

$$U_{ij}^A(r_{ij}, q_i, q_j) = -f_{S_{ij}} b_{ij} B_{ij} e^{(-\alpha_{ij} r_{ij})},$$ Short-range Attractive

$$U_{ij}^I(r_{ij}, q_i, q_j) = J_{ij}(r_{ij}) q_i q_j.$$ Electrostatic

Si_3O

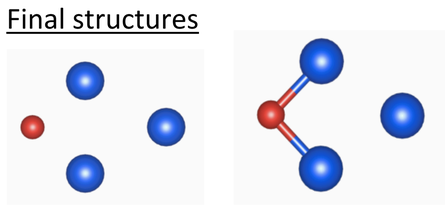
Charges

Bader	0.9363	0.9285	-0.0741	-1.7907
Pysic – pre rel.	1.7489	1.7464	-1.7935	-1.7017
Pysic – post rel.	1.9204	1.909	-2.1321	-1.6973

Bond Lengths (Å)

	VASP	Ref. [1]	ReaxFF [2]	COMB [2]	COMB (Pysic)
Si-Si	2.29	2.3	2.32	2.46	2.05
Si-O	1.72	1.75	1.69	1.72	1.54

Final structures



VASP — Pysic

Si_3O_3

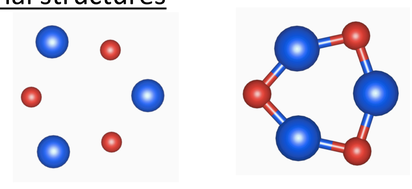
Charges

	1.5258	1.5342	1.5082	-1.5303	-1.5186	-1.5192
	1.7247	1.7247	1.7247	-1.7247	-1.7247	-1.7247
	1.6514	1.6874	1.5700	-1.6404	-1.6325	-1.6352

Bond Lengths (Å)

	VASP	Ref. [1]	ReaxFF [2]	COMB [2]	COMB (Pysic)
Si-O	1.68	1.69	1.66	1.73	1.25

Final structures



VASP — Pysic

Si_3O_4

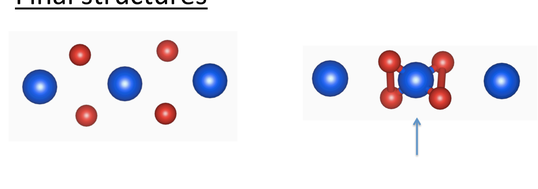
Charges

1.4139	3.0323	1.3926	-1.4695	-1.4577	-1.4479	-1.4637
1.9952	3.4242	1.9739	-1.8459	-1.8477	-1.8516	-1.8482
1.2888	4.0904	1.2685	-1.6599	-1.6624	-1.6642	-1.6611

Bond Lengths (Å)

	VASP	Ref. [1]	ReaxFF [2]	COMB [2]	COMB (Pysic)
Si-O	1.67	1.67	1.62	1.70	0.94

Final structures



VASP — Pysic

METHODS

DFT simulations performed using VASP package, with the PAW method for describing core electrons and PBE exchange correlation functional.
All simulations have been carried out in a (20X20X20) Å cubic cell, with an energy cut-off of 450 eV.
In Pysic it has been used the ASE BFGS local optimization algorithm.

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Second-generation charge-optimized many-body potential for Si/SiO_2 and amorphous silica

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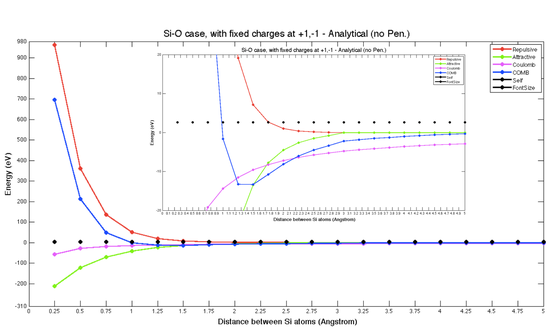
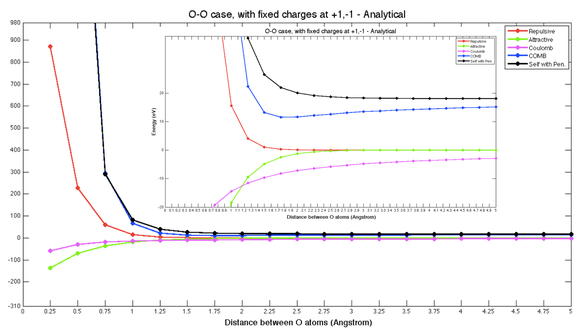
[1] Lai-Sheng Wang et al, Phys. Rev. Lett. 78, 4450–4453 (1997)
[2] Data from UCL, Alex Shluger group (2010)

COMB Trends



Si/O, with charges +1/-1 (no Penalty fc)

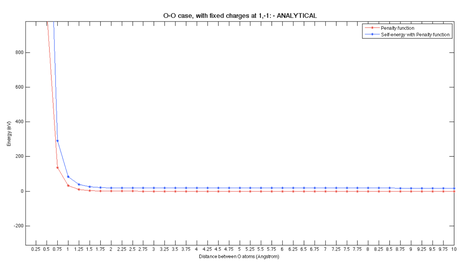


O/O, with charges +1/-1 (with Penalty fc)

Penalty Function

$$F_i^{\text{Field}}(r_{ij}, q_j) = \frac{1}{4\pi\varepsilon_0} \sum_{j\neq i}^{NN} \left(\frac{P_1^J q_j}{r_{ij}^3} + \frac{P_2^J q_j^2}{r_{ij}^5} \right)$$

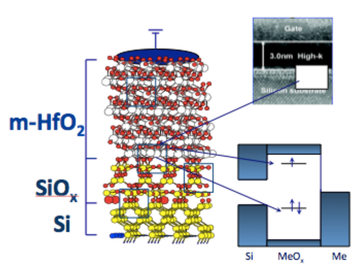
$$V_i^{Self}(r_i, q_i) = E_i^0(0) + \chi_i q_i + \left(J_i + \sum_{j\neq i}^{N} F_{ij}^{\text{Field}}(r_{ij}, q_j) \right) q_i^2 + K_i q_i^3 + L_i q_i^4$$

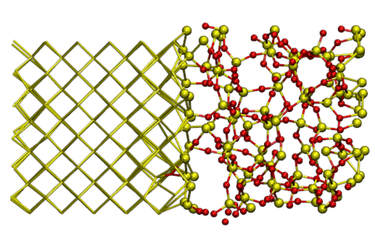


Bryce Devine et al, Phys. Rev. B 84, 125308 (2011)

Future Work

- Analysis of HfO_n (n = 2:6) clusters and crystalline polymorphs
- Simulations of new kinds of oxide/ semiconductor interfaces, like Si/HfO_2





Si/SiO_2 interface

MORDRED WP1: Interface Structure and defect properties

Contact

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