

2018.10.22

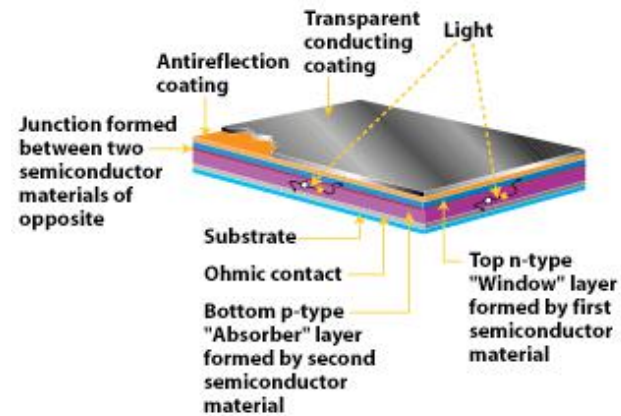
Comparison Between Cluster Expansion and MBTR for Modeling Configurational Disorder in Alloys

Xiangyue Liu, Christopher Sutton

Background | Group-III transparent conducting oxides (TCO)

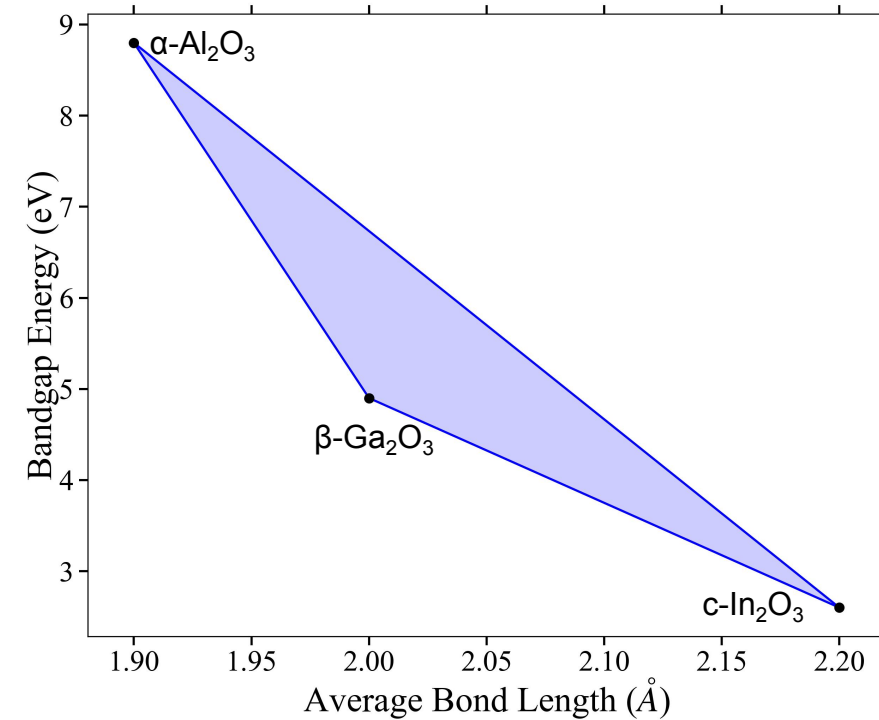
Applications:

- solar cells
- power devices
- light-emitting diodes, ...



To tune the band gaps:

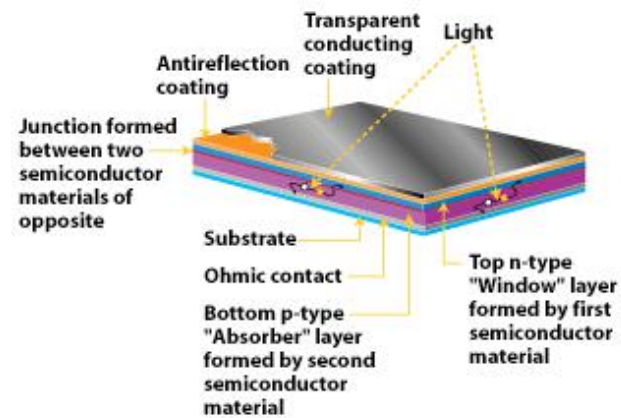
- Alloying $\text{Al}_2\text{O}_3/\text{Ga}_2\text{O}_3/\text{In}_2\text{O}_3$
- Including configurational disorders



Background | Group-III transparent conducting oxides (TCO)

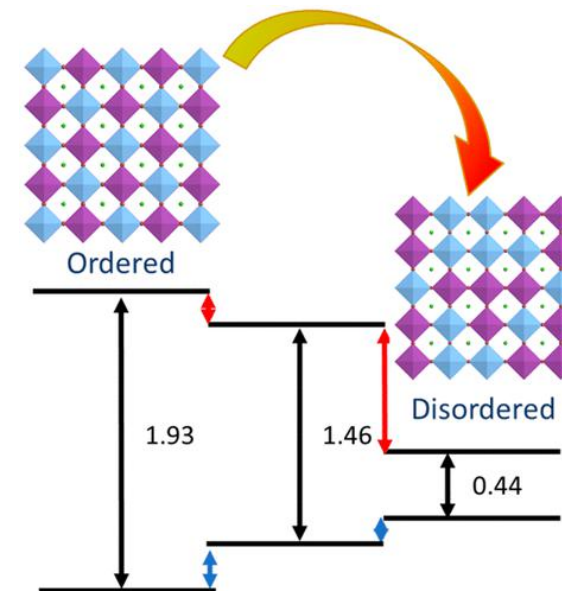
Applications:

- solar cells
- power devices
- light-emitting diodes, ...



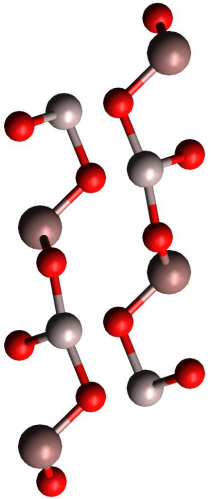
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- Alloying $\text{Al}_2\text{O}_3/\text{Ga}_2\text{O}_3/\text{In}_2\text{O}_3$
- Including configurational disorder

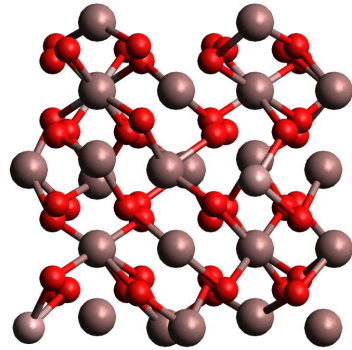


Background | Configurational disorder in $(\text{Al}_x\text{Ga}_y\text{In}_{1-x-y})_2\text{O}_3$ alloys

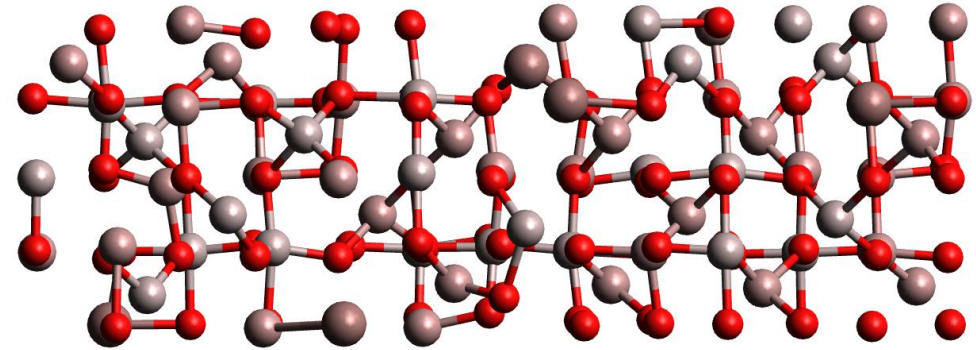
C2/m (12)



Ia-3 (206)



Fd-3m (227)



Background | Configurational disorder in $(\text{Al}_x\text{Ga}_y\text{In}_{1-x-y})_2\text{O}_3$ alloys

Fd-3m structure:

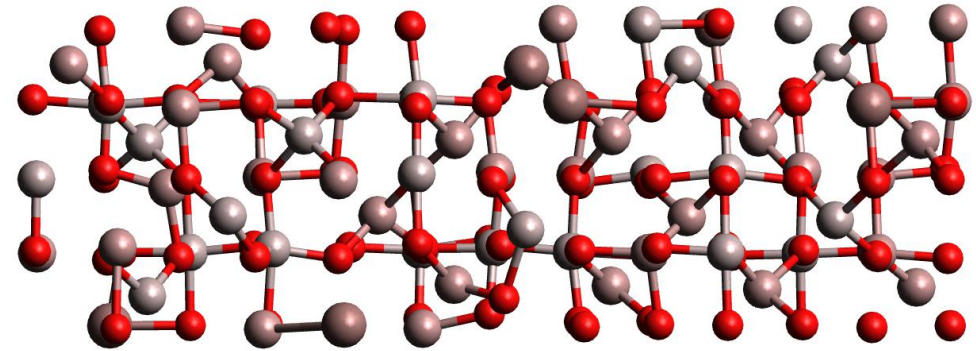
cation:anion = 3:4

M_2O_3 :

cation:anion = 2:3

- > Vacancy defects on the cation sublattice
- > Inherent structural disorder
- > Structural distortion

Fd-3m (227)

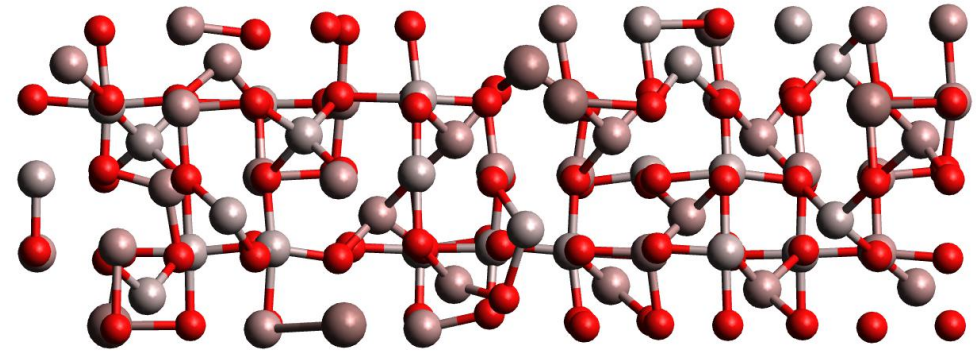


Background | $(\text{Al}_x\text{Ga}_y\text{In}_{1-x-y})_2\text{O}_3$ alloys: Model transferability to doped systems?

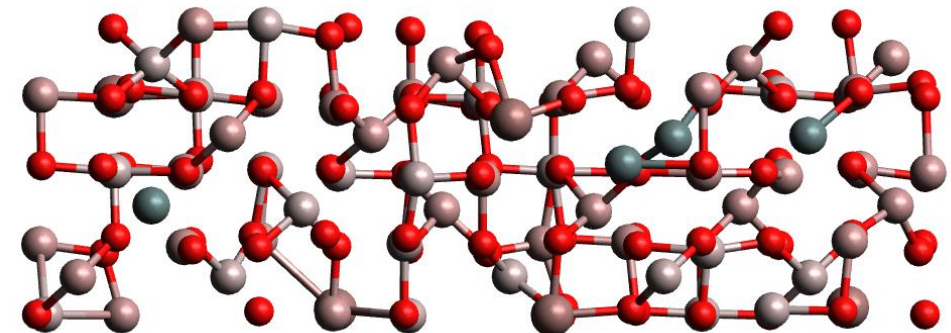
Sn(II)-doped Fd-3m:

> Model transferability from undoped to doped alloys?

Fd-3m (227)

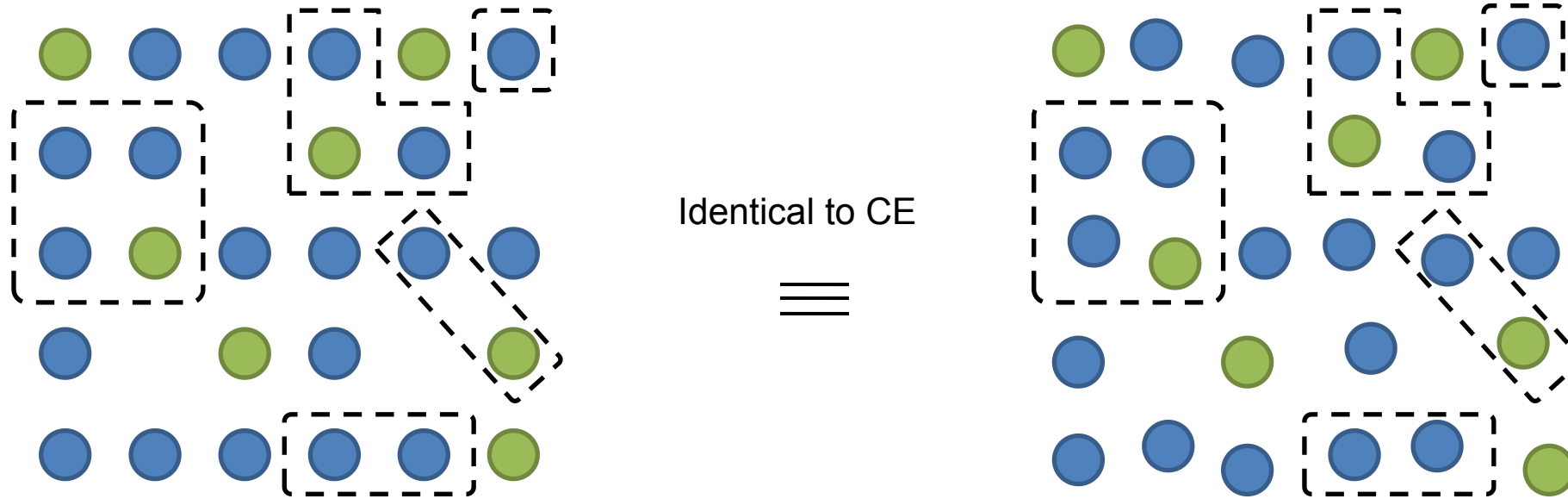




Sn(II)-doped Fd-3m



Background | Cluster expansion (CE): when it may fail?

No information about interatomic distance/angles -> Structural distortions can not be taken into account



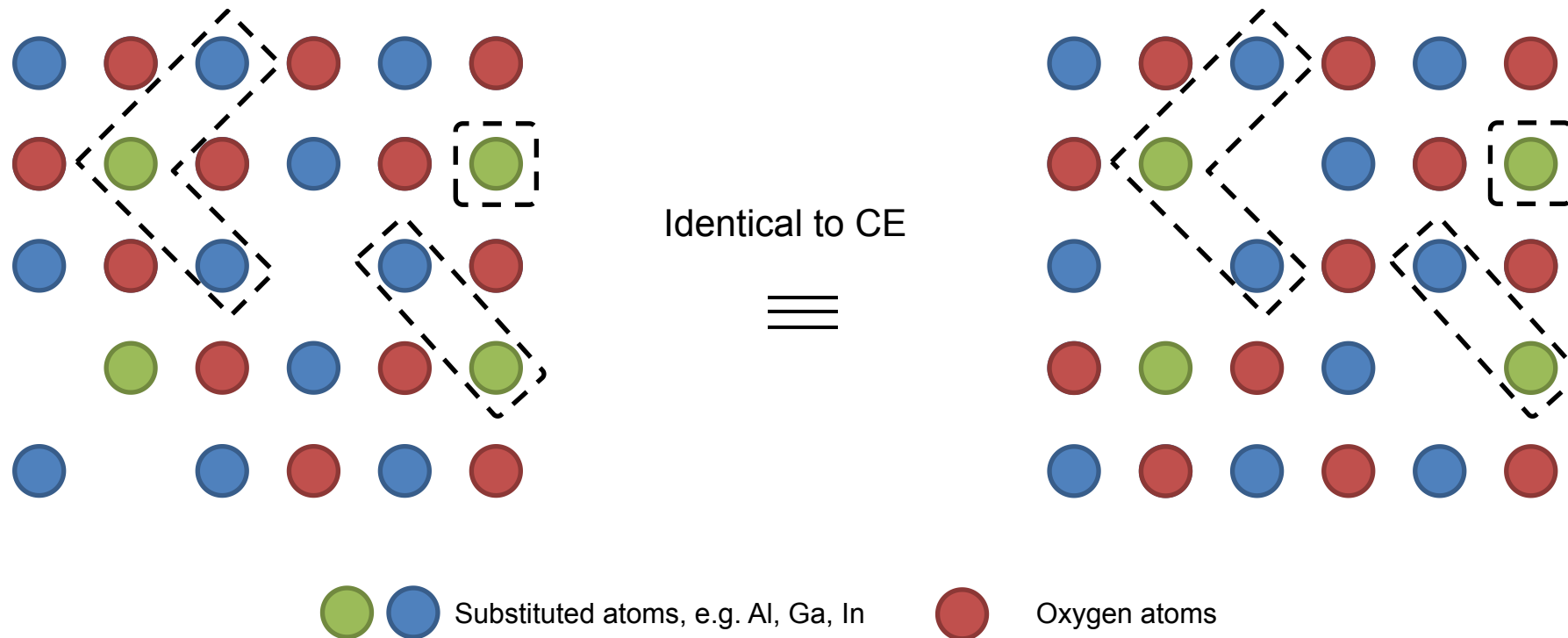
  Substituted atoms, e.g. Al, Ga, In

$$E(\sigma) = \sum_{\alpha} J_{\alpha} X_{\alpha}$$



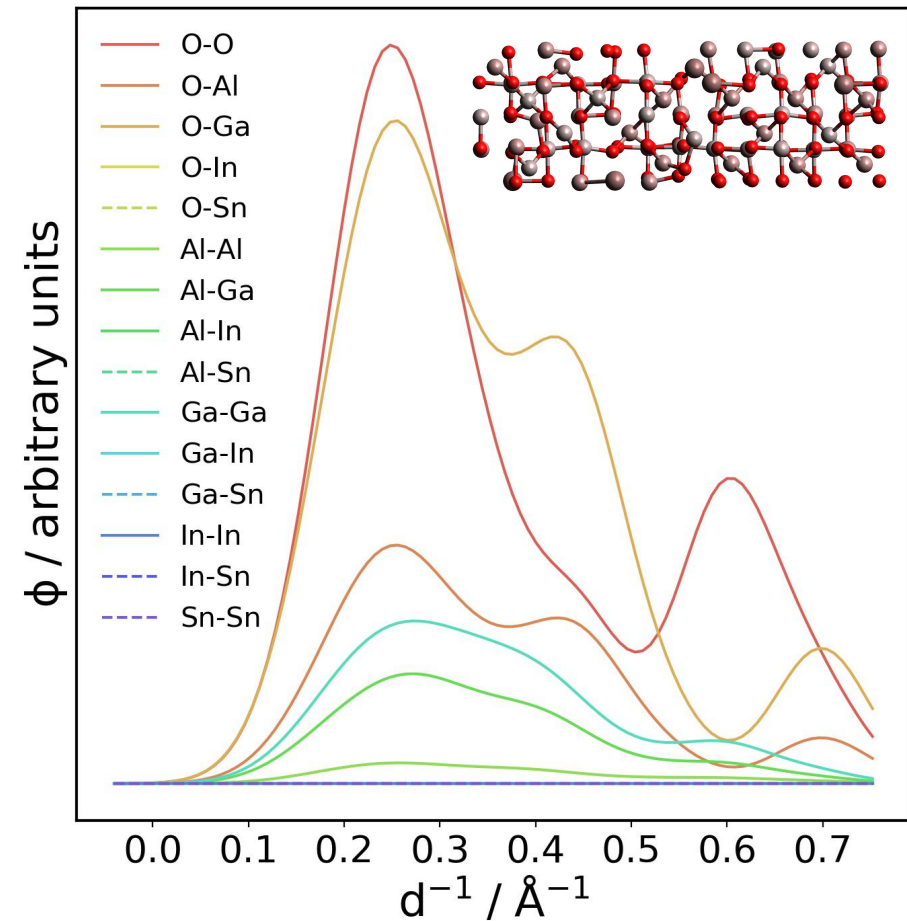
Background | Cluster expansion (CE): when it may fail?

Only substituted atoms are included in the basis: e.g. for $(\text{Al}_x\text{Ga}_y\text{In}_{1-x-y})_2\text{O}_3$ alloys only metal-metal clusters



Background | Many-body tensor representation (MBTR)

$$f_k(x, \mathbf{z}) = \sum_{i=1}^{N_a} w_k(\mathbf{i}) D(x, g_k(\mathbf{i})) \prod_{j=1}^k C_{z_j, Z_{i_j}}$$

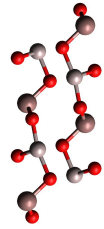


CE predictions of C2/m and Ia-3 ($\text{Al}_x\text{Ga}_y\text{In}_{1-x-y}\text{O}_3$) alloys

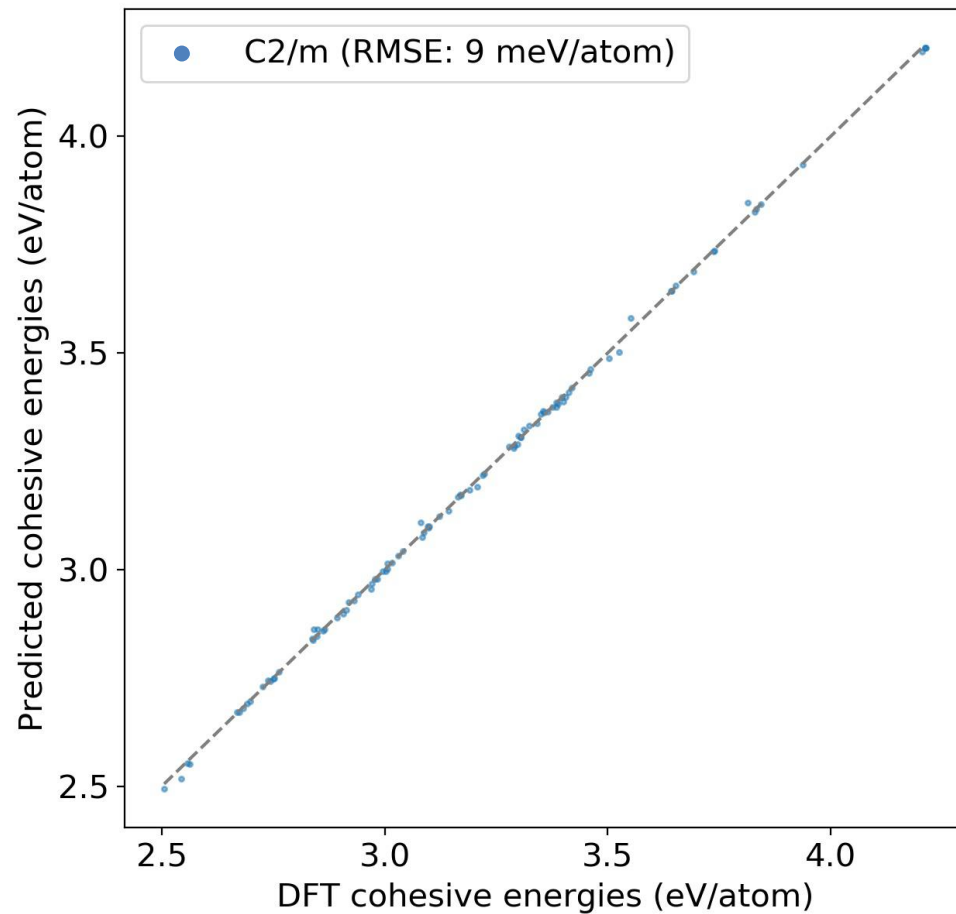
C2/m

Training set: 358 alloys

Test set: 97 alloys



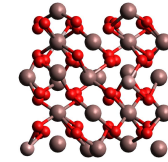
PBE, "tight"



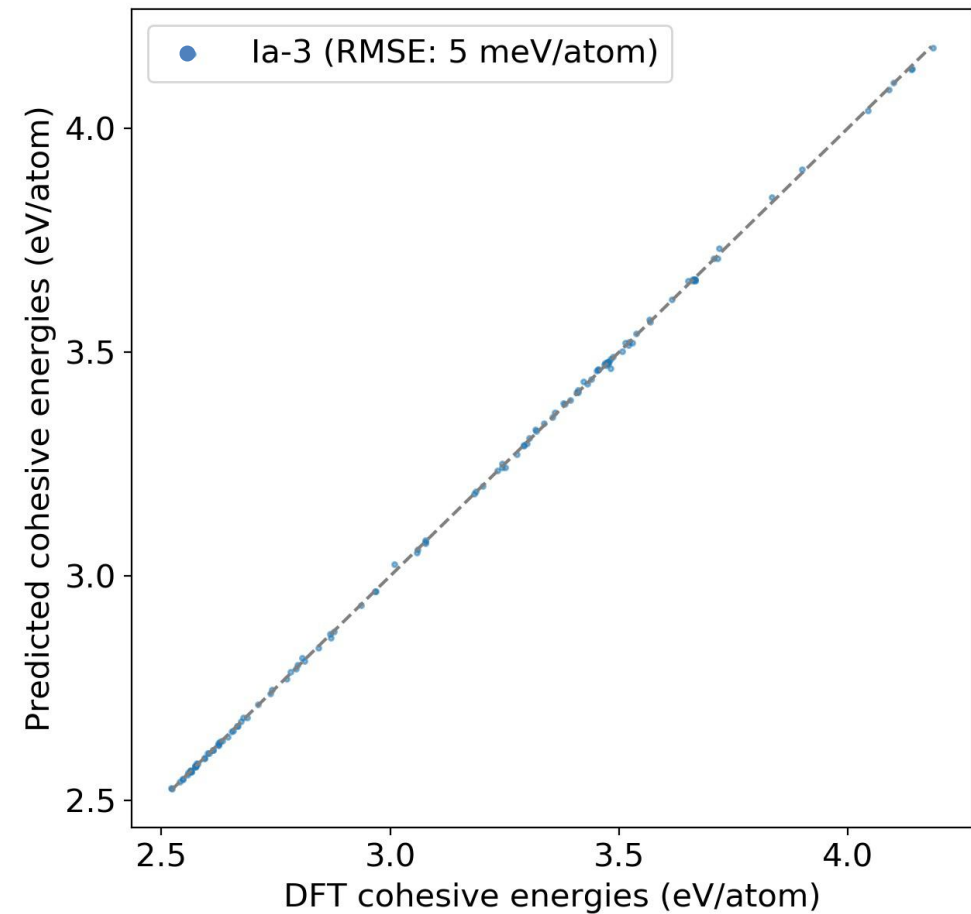
Ia-3

Training set: 490 alloys

Test set: 126 alloys



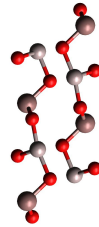
PBE, "tight"



MBTR+KRR predictions of C2/m and Ia-3 ($\text{Al}_x\text{Ga}_y\text{In}_{1-x-y}\text{O}_3$) alloys

C2/m

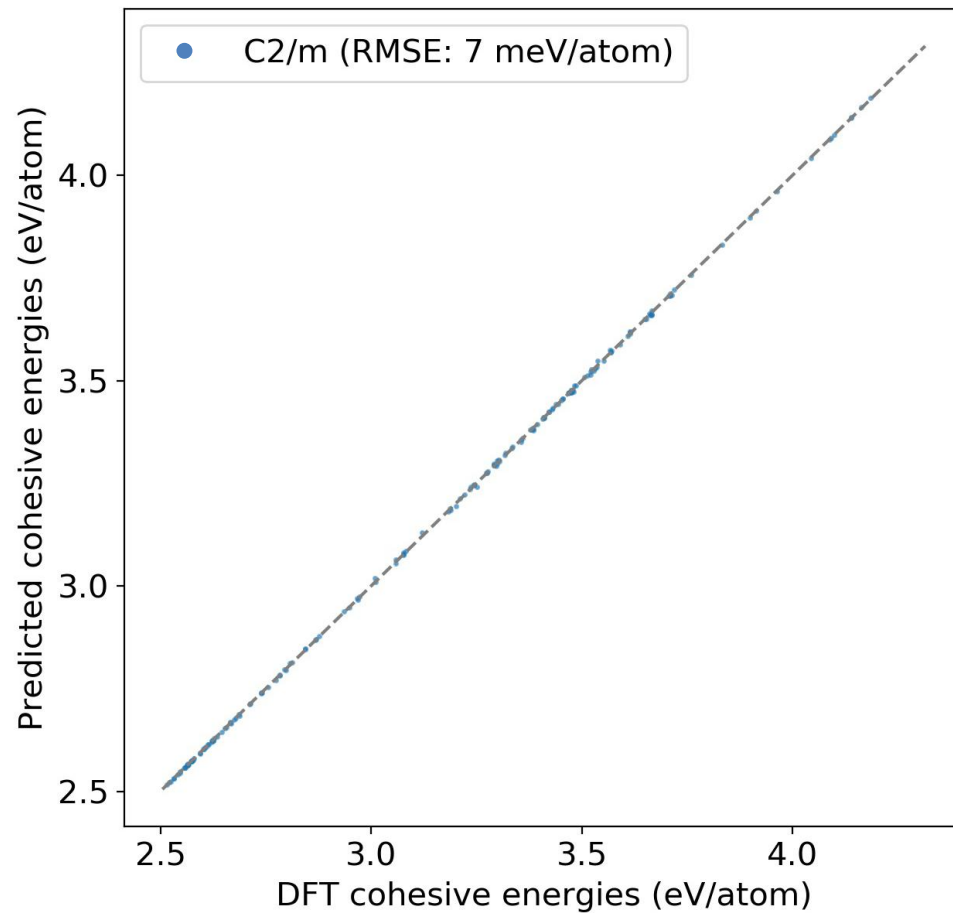
Vegard's law structures as inputs



PBE, "tight"

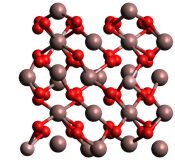
Training set: 320 alloys

Test set: 135 alloys



Ia-3

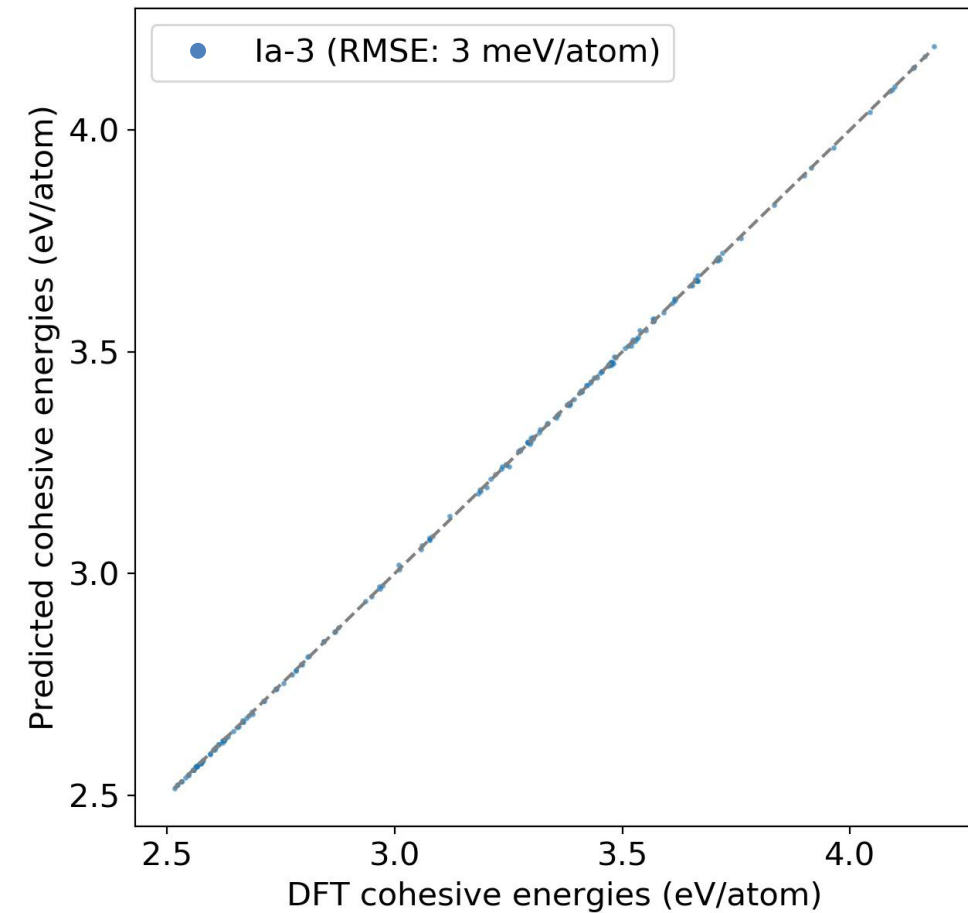
Vegard's law structures as inputs



PBE, "tight"

Training set: 430 alloys

Test set: 186 alloys



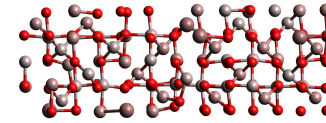
Predictions of Fd-3m $(\text{Al}_x\text{Ga}_y\text{In}_{1-x-y})_2\text{O}_3$ alloys: CE vs MBTR+KRR

Undoped Fd-3m

Training set: 1400 undoped Fd-3m alloys

Test set: 600 undoped Fd-3m alloys

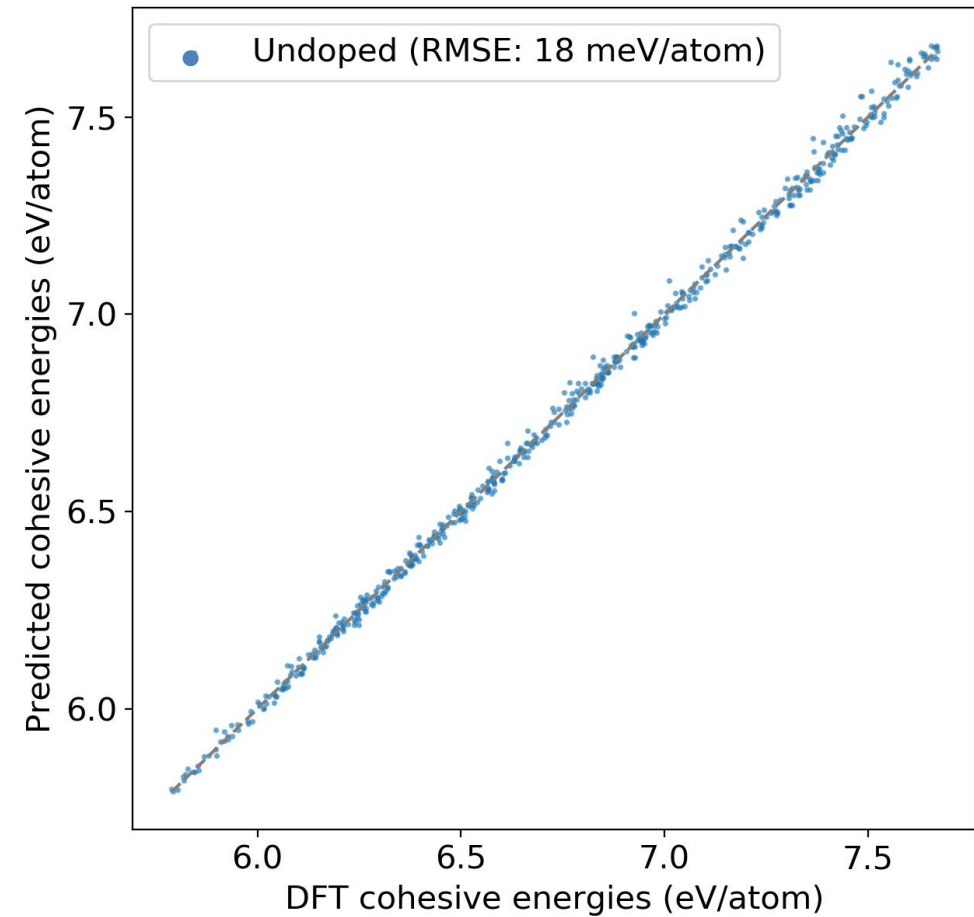
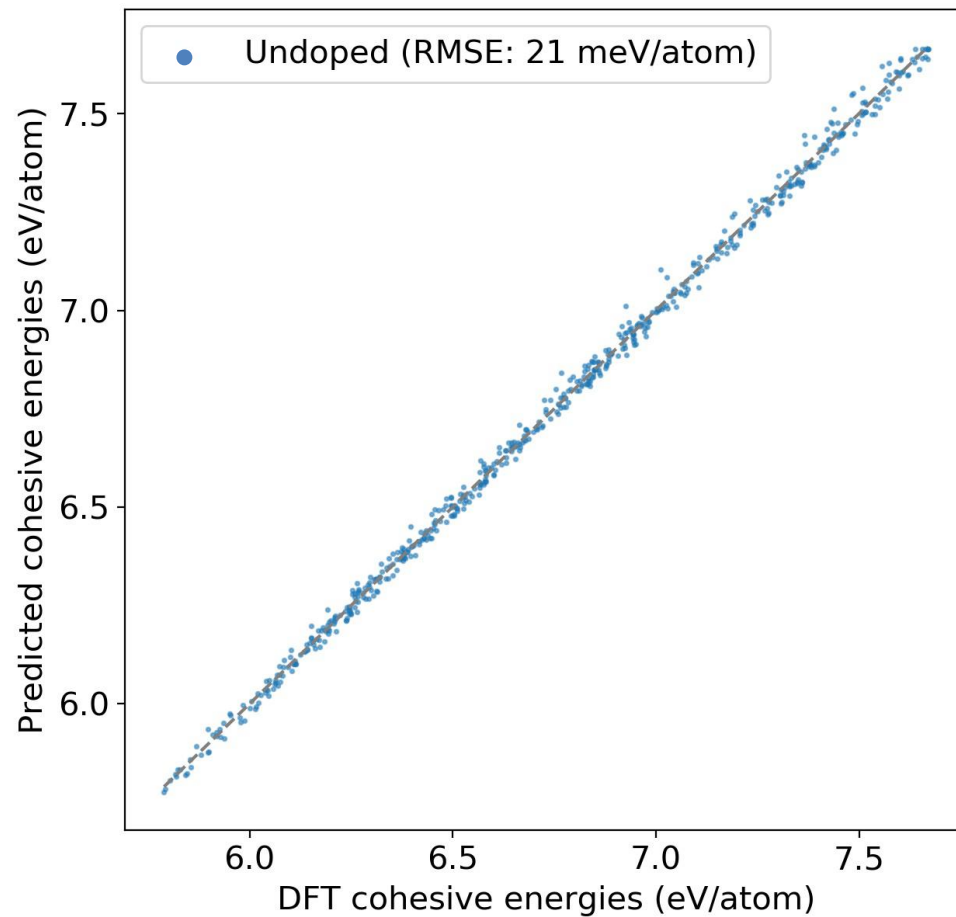
160 atoms/cell, PBEsol, "light"



CE

MBTR

Vegard's law structures as inputs



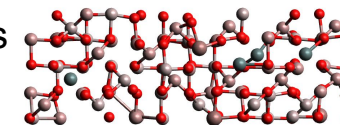
Transferability from undoped to doped alloys: CE vs MBTR+KRR

Undoped + Sn(II)-doped Fd-3m

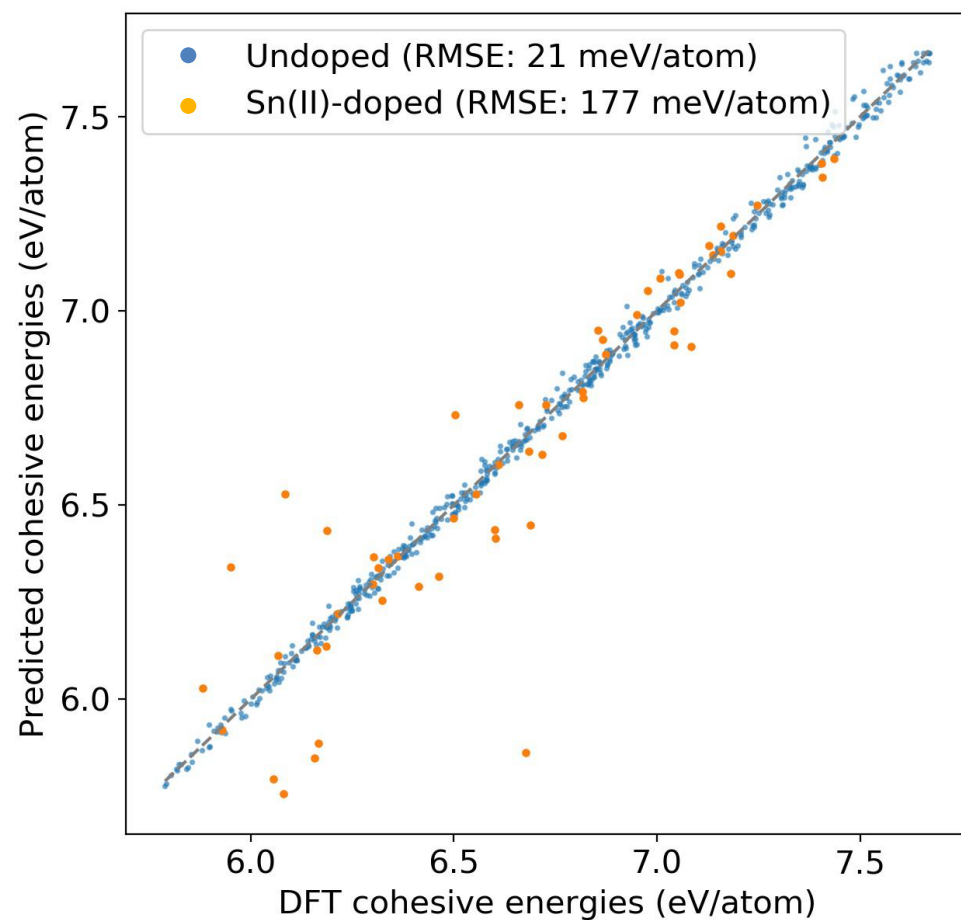
Training set: 1400 undoped Fd-3m alloys + 140 Sn(II)-doped Fd-3m alloys

Test set: 600 undoped Fd-3m alloys + 58 Sn(II)-doped Fd-3m alloys

160 atoms/cell, PBEsol, "light"

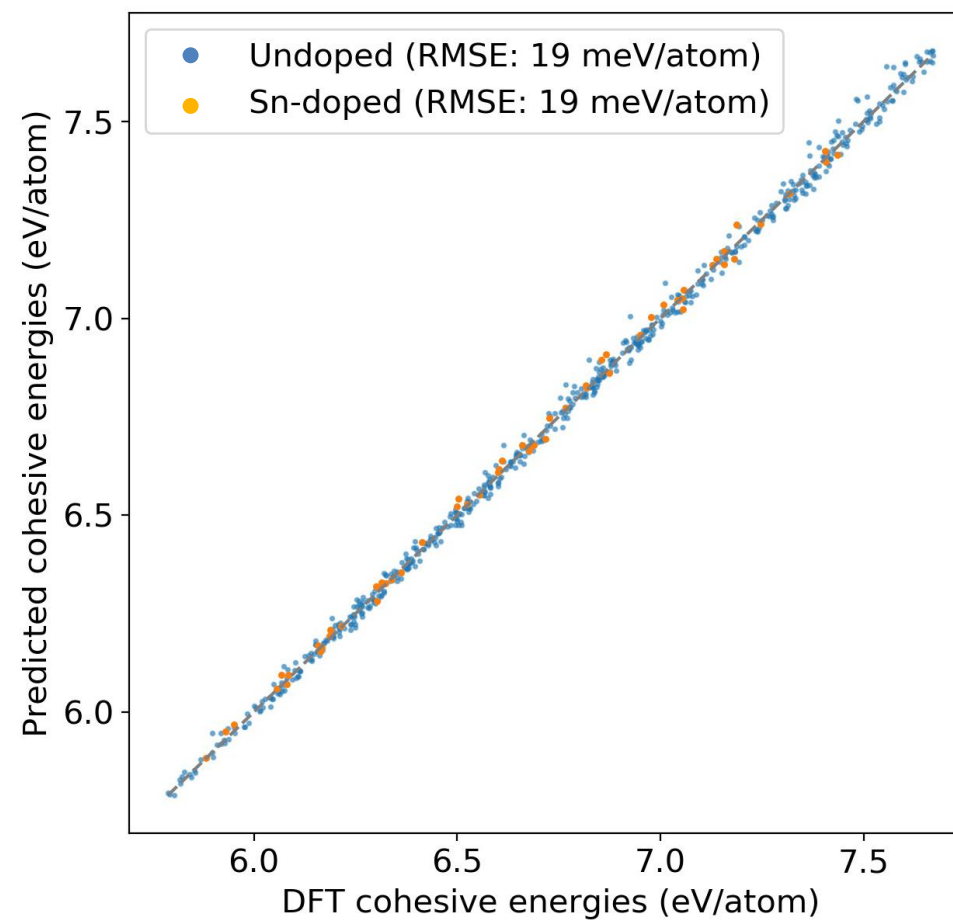


CE



MBTR

Vegard's law structures as inputs

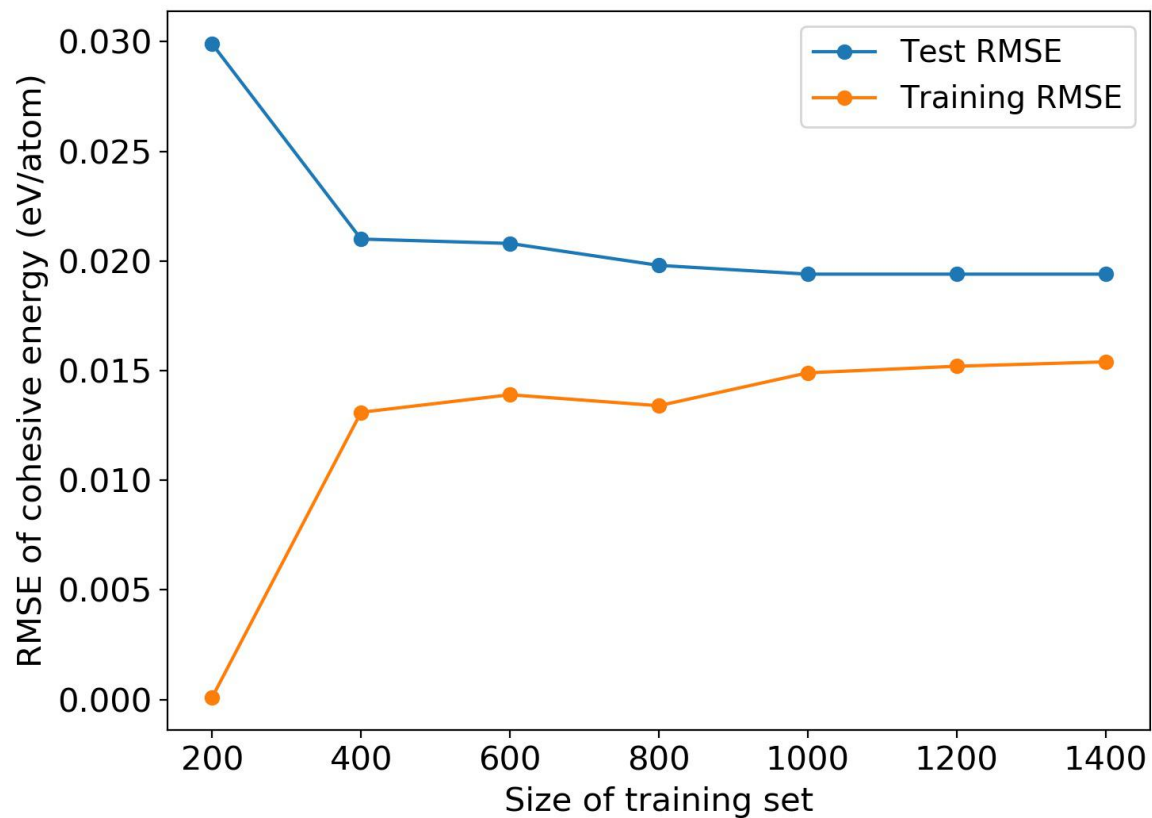


MBTR+KRR : Learning curves: undoped v.s. doped

Undoped Fd-3m

Training set: 1400 undoped Fd-3m alloys

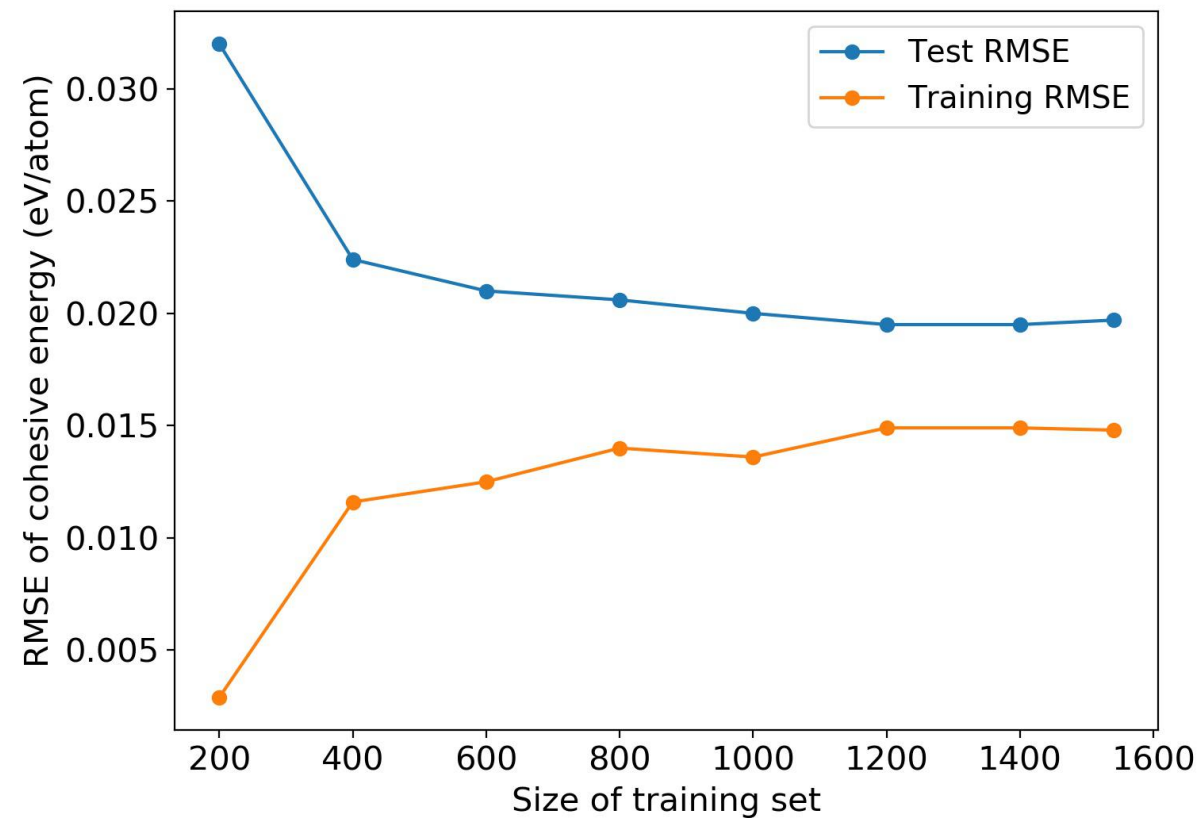
Test set: 600 undoped Fd-3m alloys



Undoped + Sn(II)-doped Fd-3m

Training set: 1400 undoped Fd-3m alloys + 140 Sn-doped Fd-3m alloys

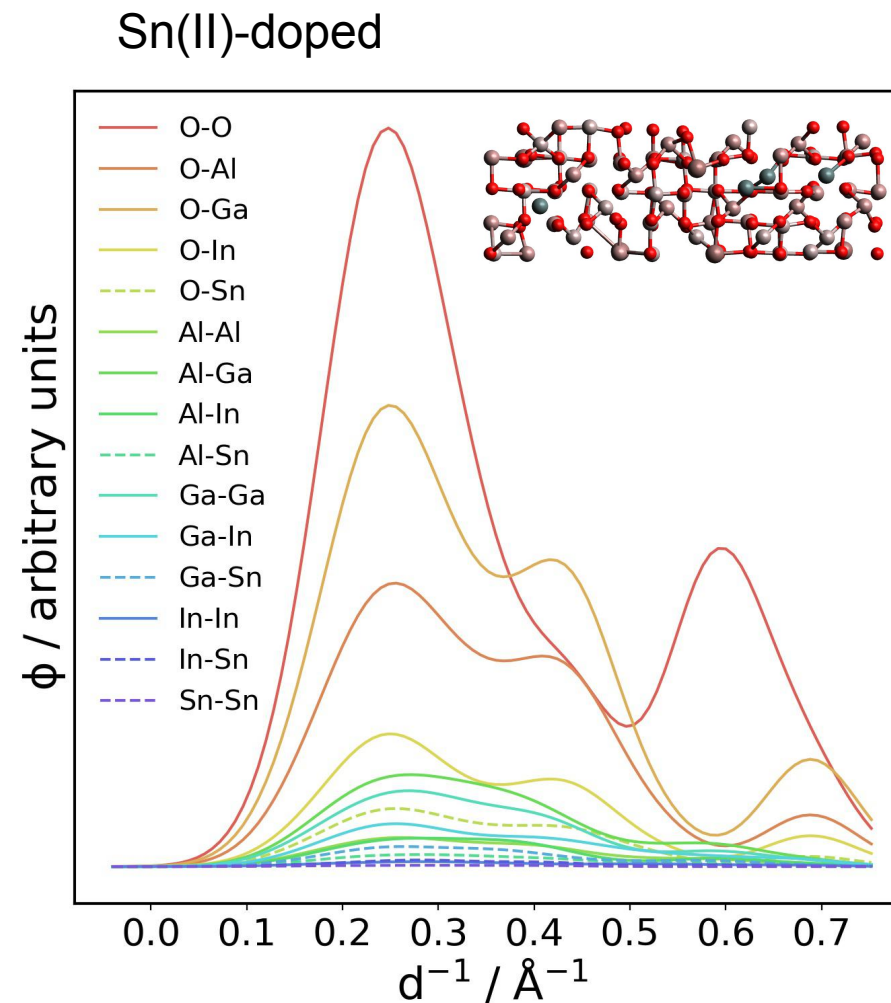
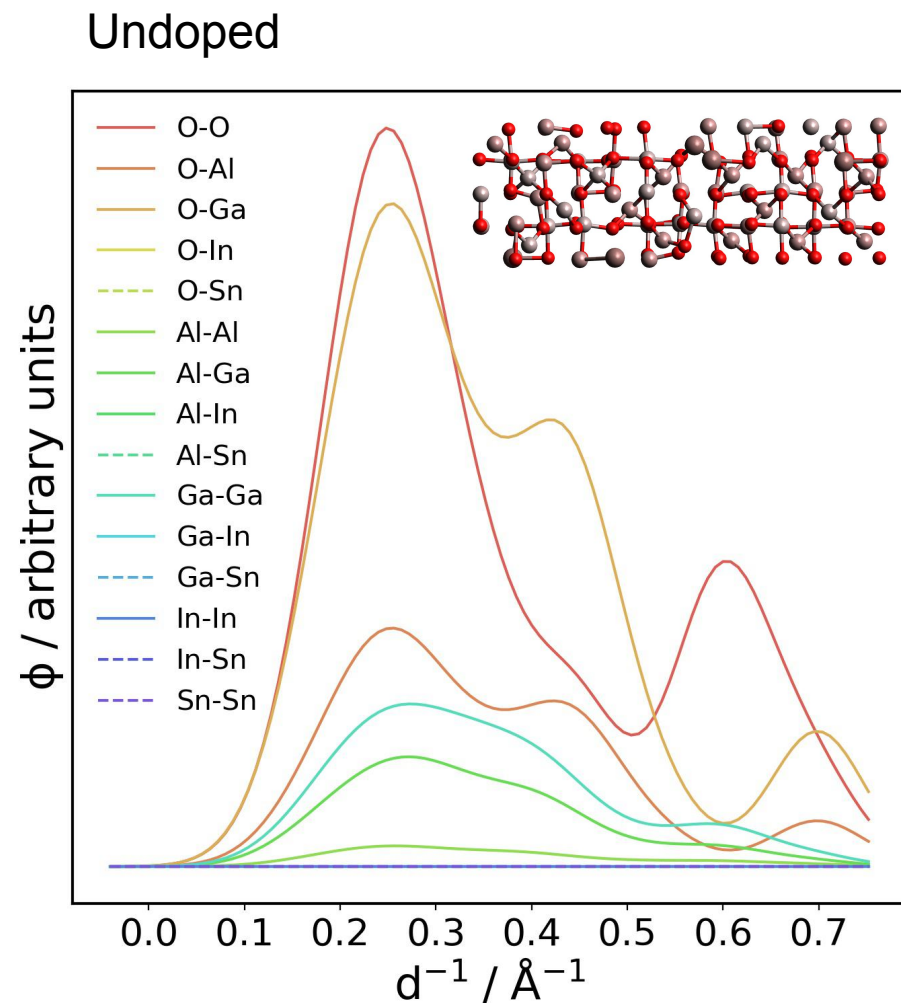
Test set: 600 undoped Fd-3m alloys + 58 Sn-doped Fd-3m alloys



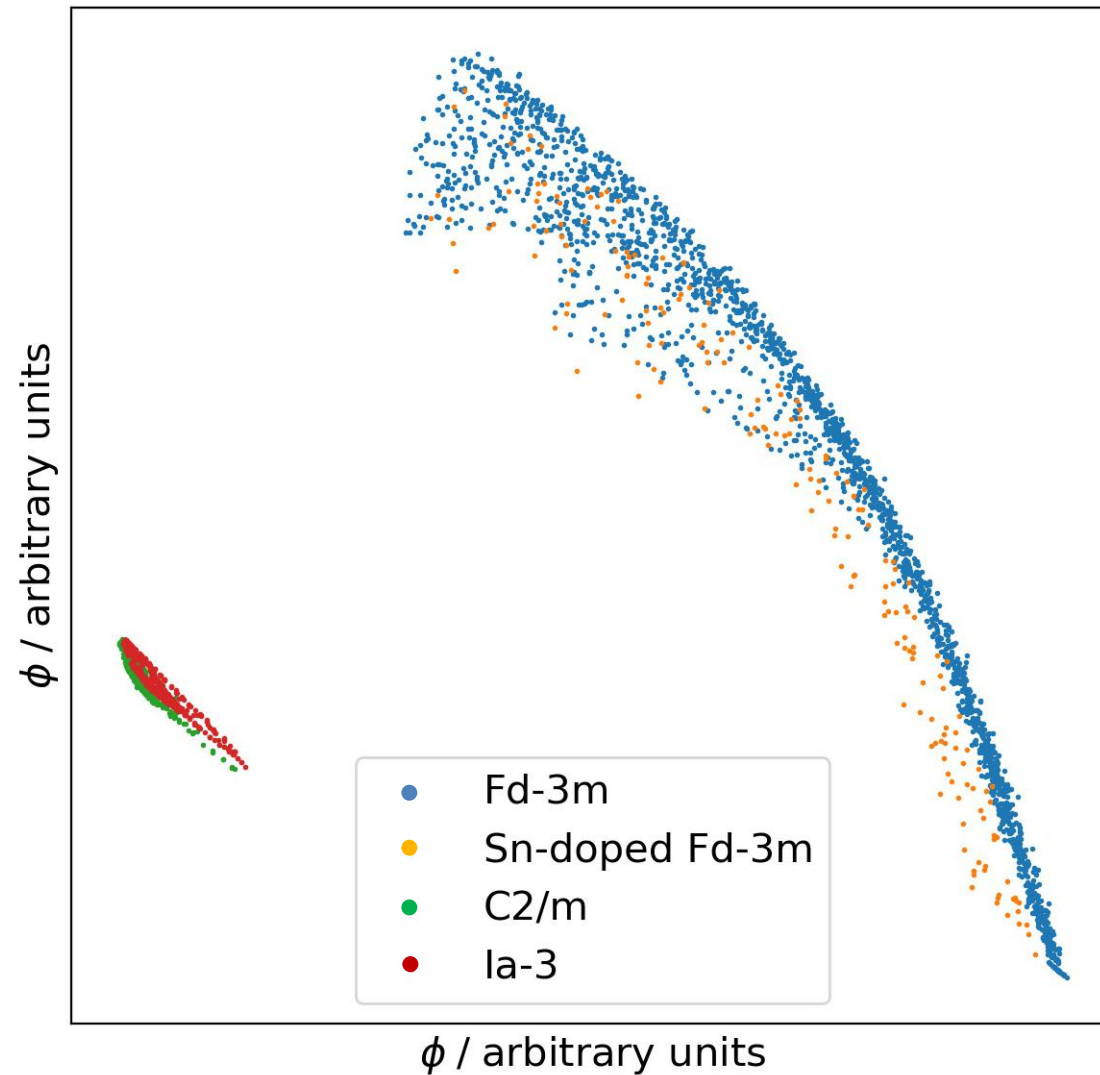
MBTR functions: transferability from undoped to doped alloys

Training set: 1400 undoped Fd-3m alloys + 140 Sn-doped Fd-3m alloys

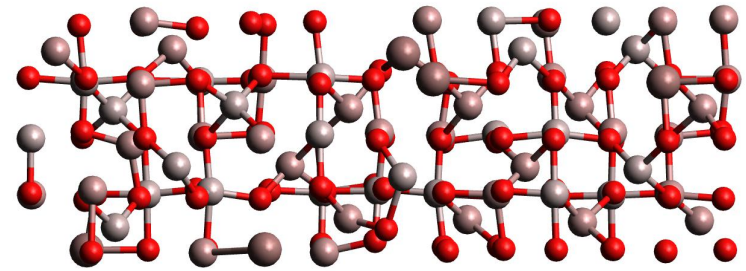
Test set: 600 undoped Fd-3m alloys + 58 Sn-doped Fd-3m alloys



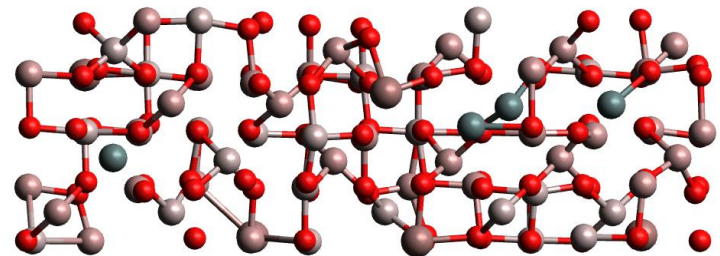
Principal components analysis (PCA)



Undoped



Sn(II)-Doped



Summary

	CE	MBTR+KRR
Modeling C2/m, Ia-3, Fd-3m independently	✓	✓
Transferability from undoped to doped alloys		✓

Acknowledgements

Christopher Sutton
Marcel Langer
Matthias Rupp

Thanks for attention.