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Computing Grain Boundary Diagrams for High-Entropy Alloys

PRESENTED BY

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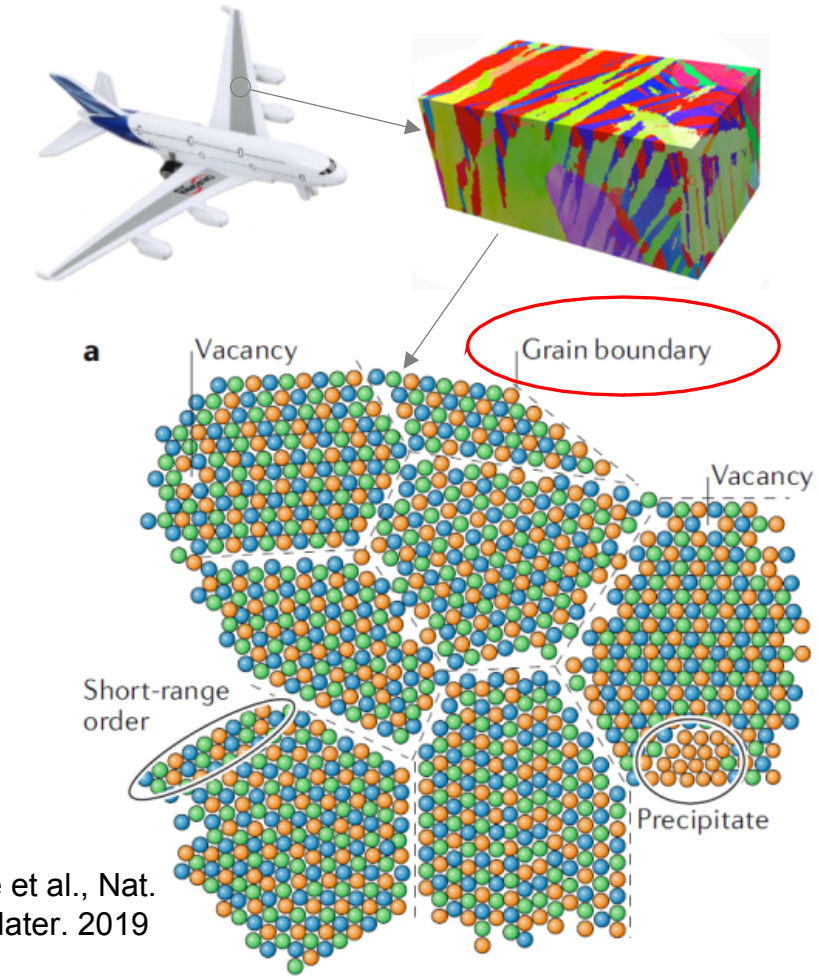
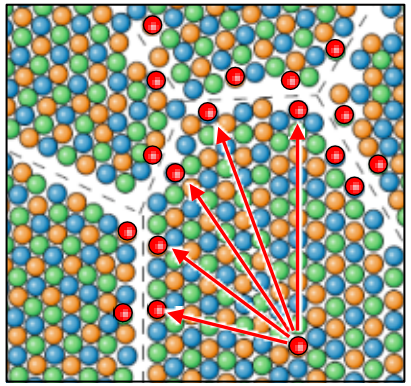
Background

Grain boundaries (GBs) are one of the most common crystal defects in polycrystalline materials



George et al., Nat. Rev. Mater. 2019

GBs can interact with foreign atoms (red below) and results in GB segregation

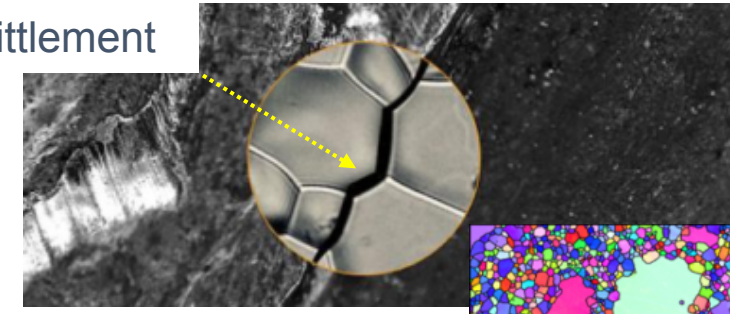


Host elements

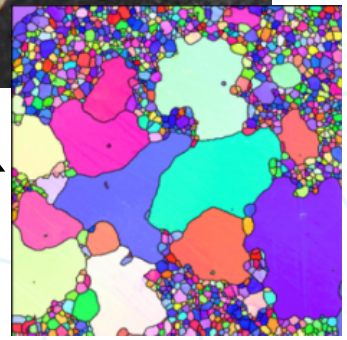
Solute (Impurity) atoms

Even a tiny solute (or impurity) segregation

GB embrittlement



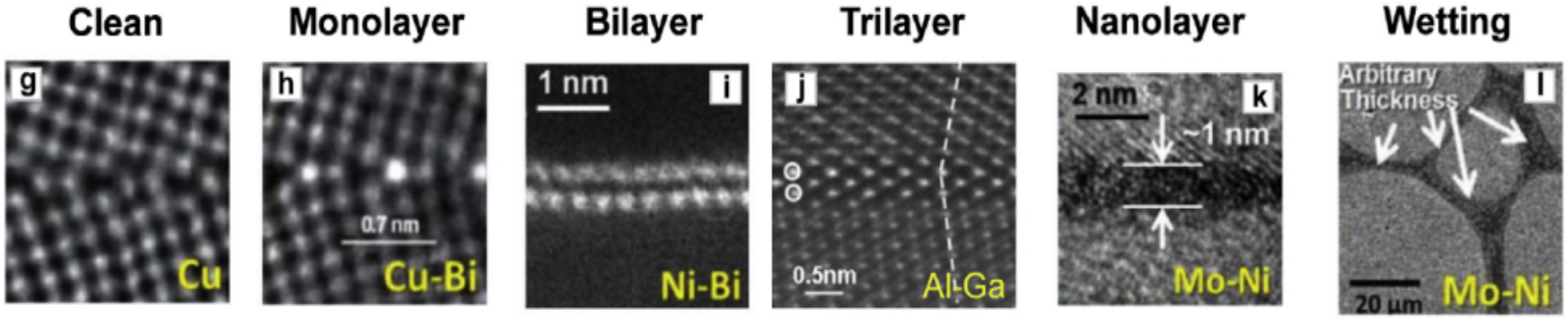
Abnormal grain growth



Many other properties

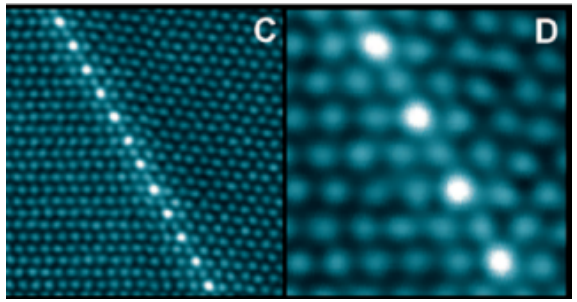
Background

Segregation has been broadly studied for many GB structures in different materials



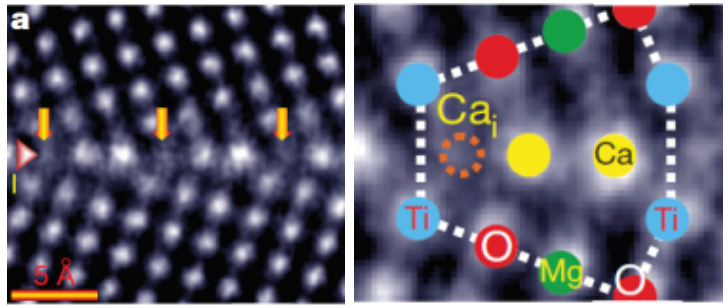
Luo et al., Science 2011

Periodic monolayer segregation of solute atoms in Mg TB



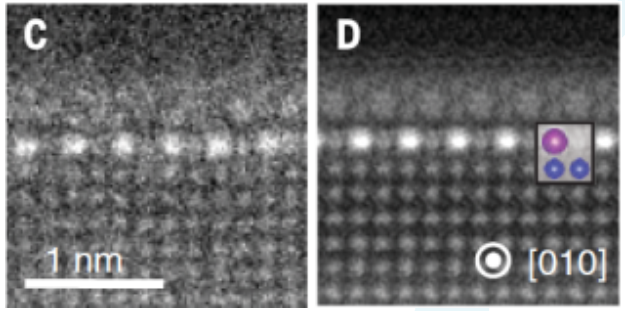
Nie et al., Science 2013

Ordered superstructure in MgO Σ3 GB



Wang et al., Nature 2011

Bi bilayer superstructure in Ni Polycrystals



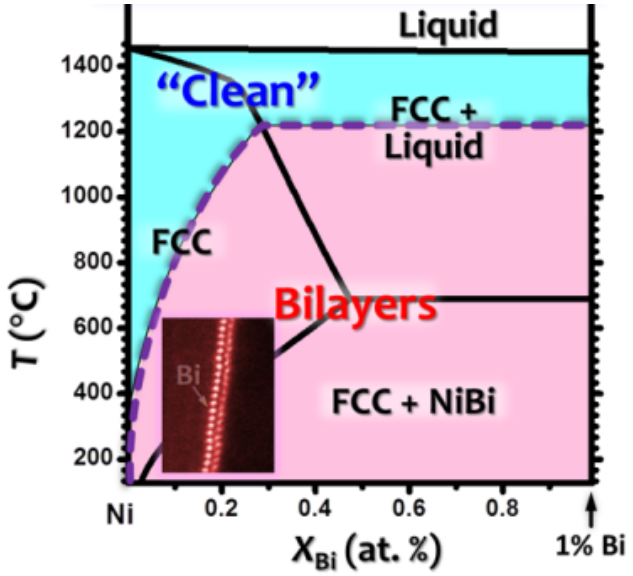
Yu et al., Science 2018

However, experiments are complicate and time consuming. Only few thermodynamic conditions (at certain temperature and composition) were studied

Background

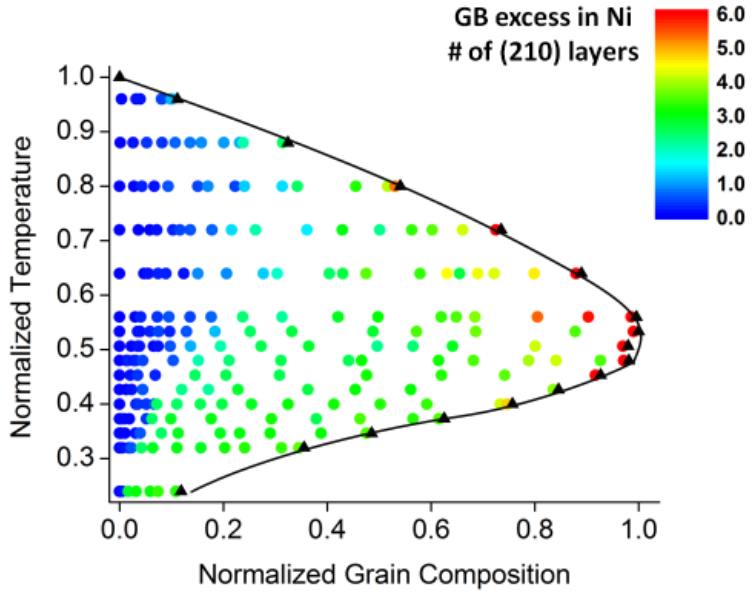
Develop grain boundary diagrams is one of effective methods to study segregation as a function of temperature and composition

Ni-Bi Adsorption Diagram



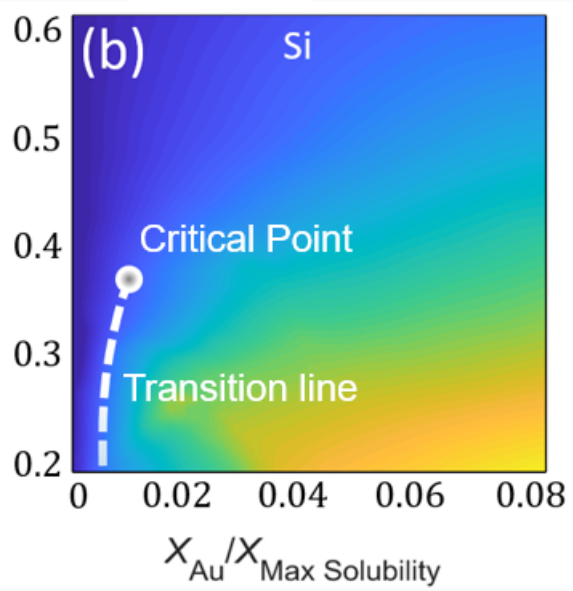
Zhou et al., Scripta 2017

Mo-Ni Adsorption Diagram



Luis et al., Nature 2017

Si-Au Adsorption Diagram



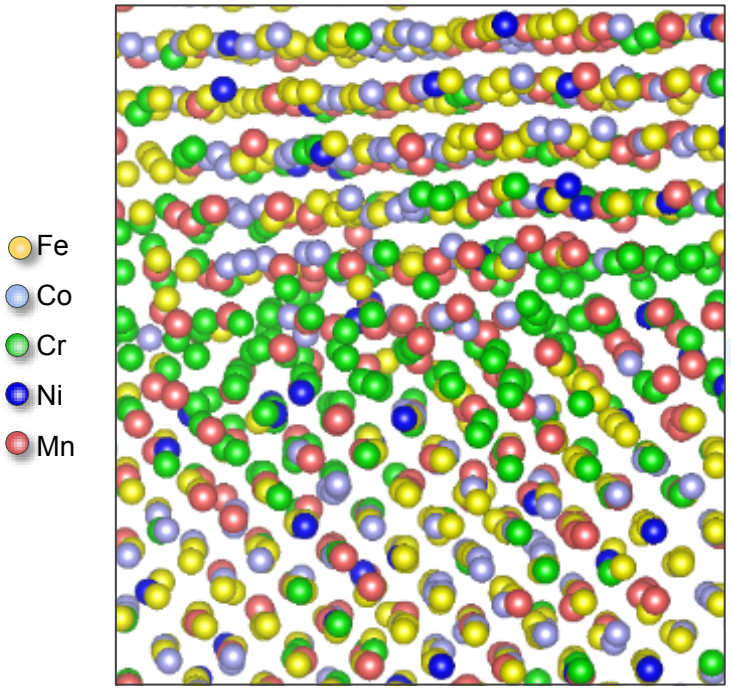
Hu et al., Scripta 2019

However, most of these GB diagrams are developed for binary alloys with one compositional degrees of freedom

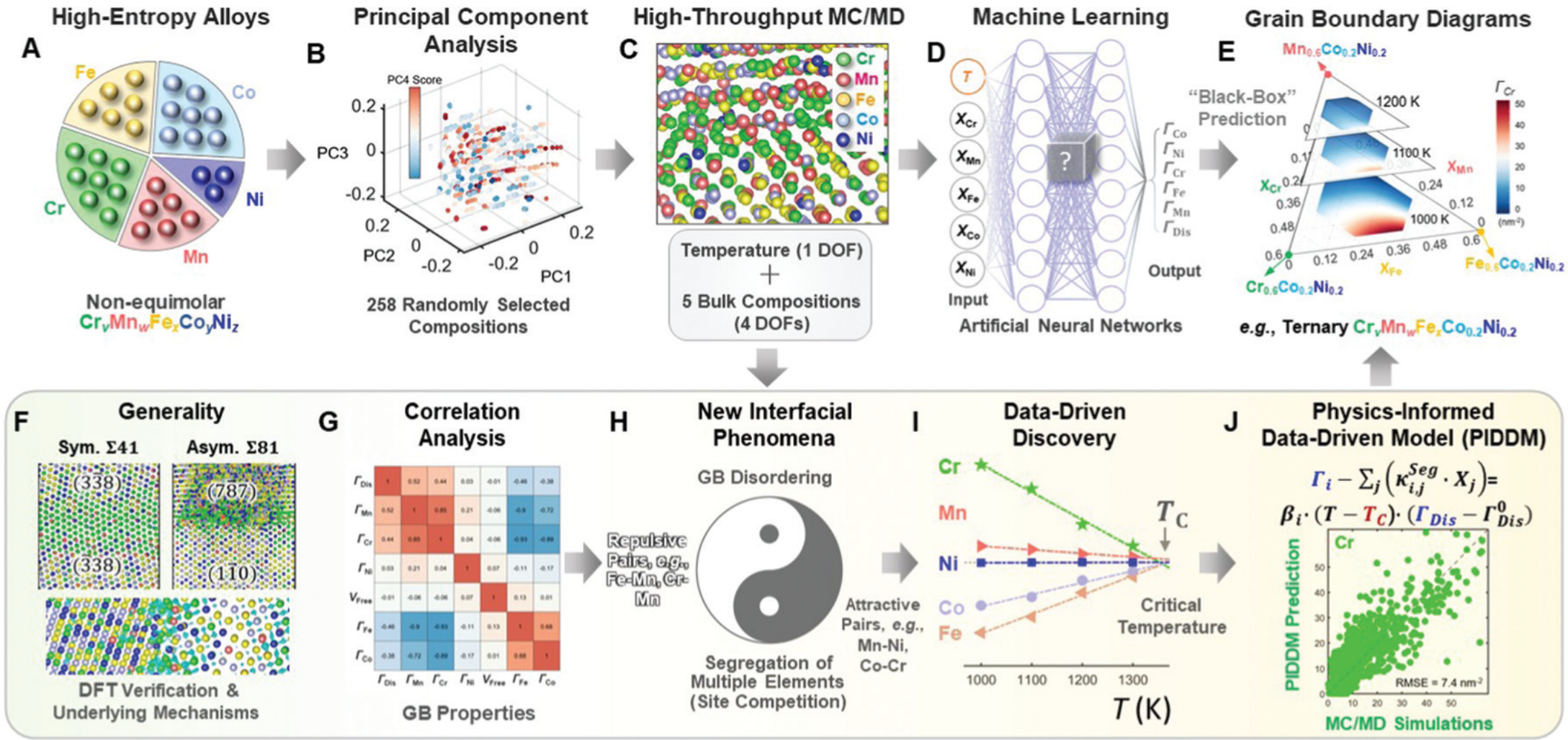
Motivation and Method

- High-entropy alloys (HEAs) generally has five or more elements, leading to a large compositional space
- Using CrMnFeCoNi (Cantor alloy) as model system to compute GB diagrams for HEAs
- Applied hybrid Monte Carlo/molecular dynamic (MC/MD) simulations to develop large datasets for 258 different compositions at 4 different temperatures
- At each condition, we compute five GB adsorption properties and disorder for an asymmetric Σ81 GB (represent a general GB)
- The large GB dataset was used to train several machine learning models
- A physics-informed data-based model was developed to predict GB properties of HEAs

GB Structure of CrMnFeCoNi



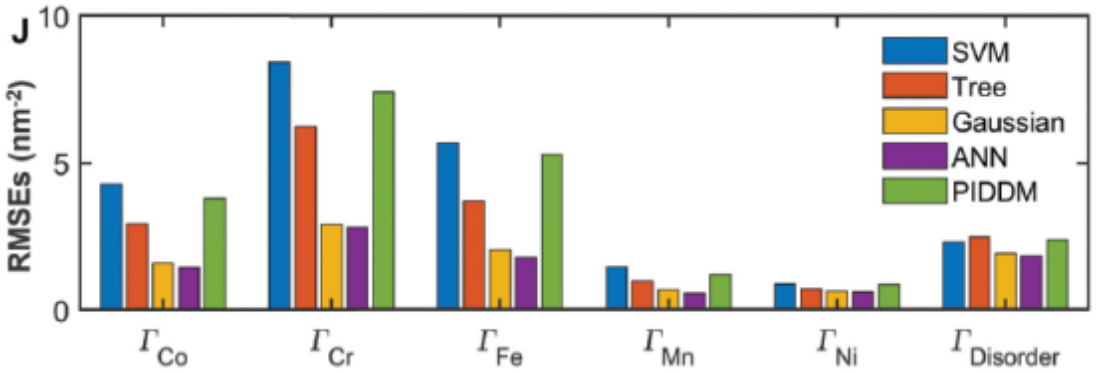
Workflow of Data-Driven Prediction of GB Properties of HEAs

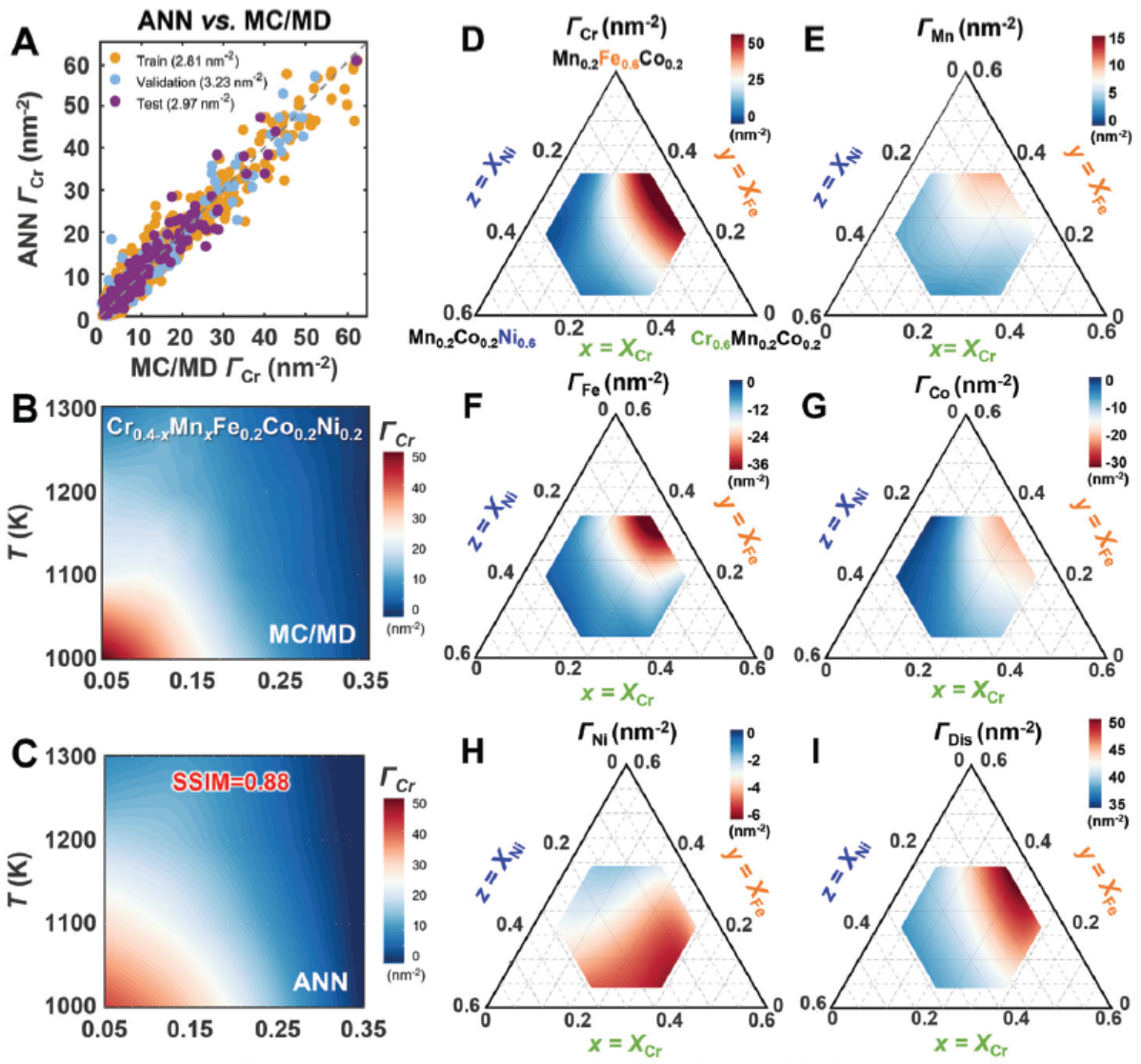


Hu and Luo, Materials Horizons, 2022

Machine Learning Prediction of Grain Boundary Diagrams

- Artificial neural network (ANN) is the most accurate model to predict GB properties among four machine learning models
- ANN-predicted GB adsorption has good agreement with MC/MD simulations
- For instance, ANN-predicted Cr adsorption diagram in $Cr_{0.4-x}Mn_xFe_{0.2}Co_{0.2}Ni_{0.2}$ is consistent with MC/MD simulations
- ANN models are more effective to predict ternary GB diagrams

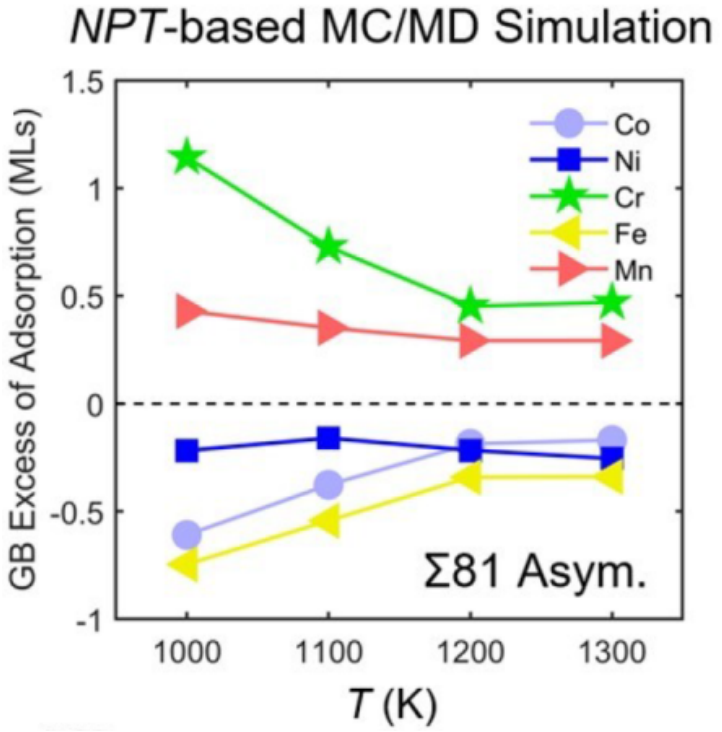


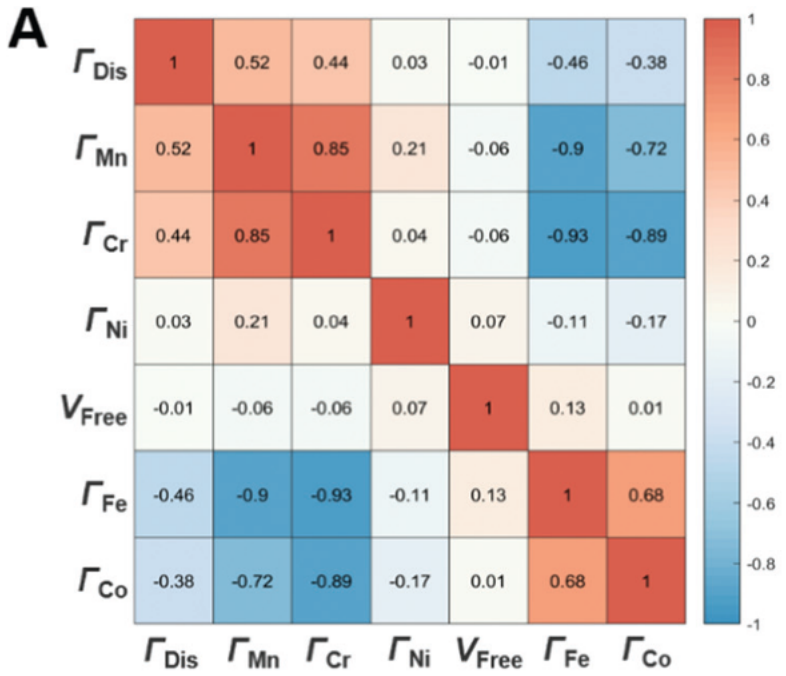


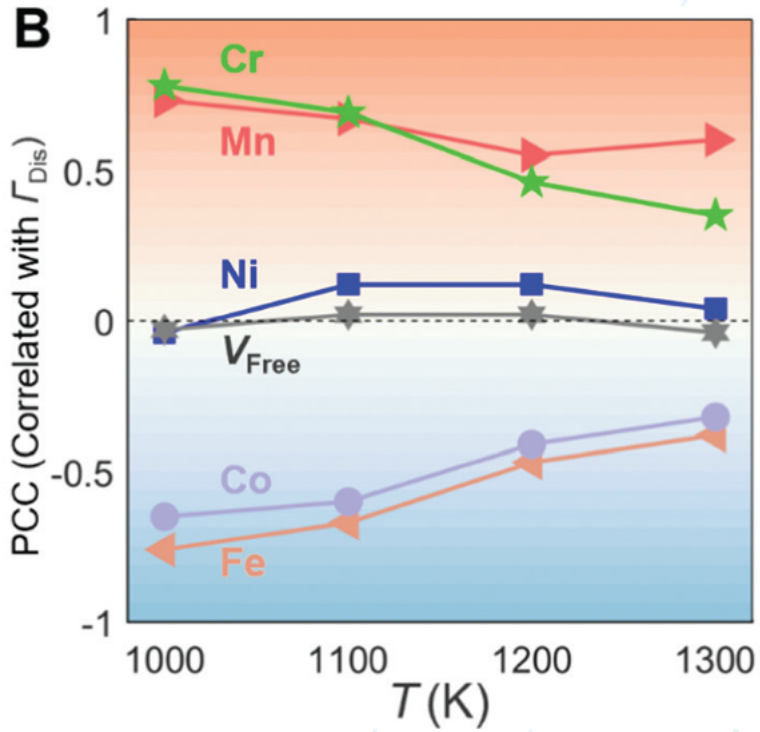
Hu and Luo, Materials Horizons, 2022

Correlation Analysis for GB Properties

- GB segregation trend: $\Gamma_{Cr} > \Gamma_{Mn} > 0 > \Gamma_{Ni} > \Gamma_{Co} \sim \Gamma_{Fe}$ (Cr and Mn: segregation, Co and Fe: GB depletion)
- GB excess of disorder (Γ_{Dis}) has strong positive correlation with Cr (Γ_{Cr}) and Mn (Γ_{Mn}), but negative correlation with Co (Γ_{Co}) and Fe (Γ_{Fe})

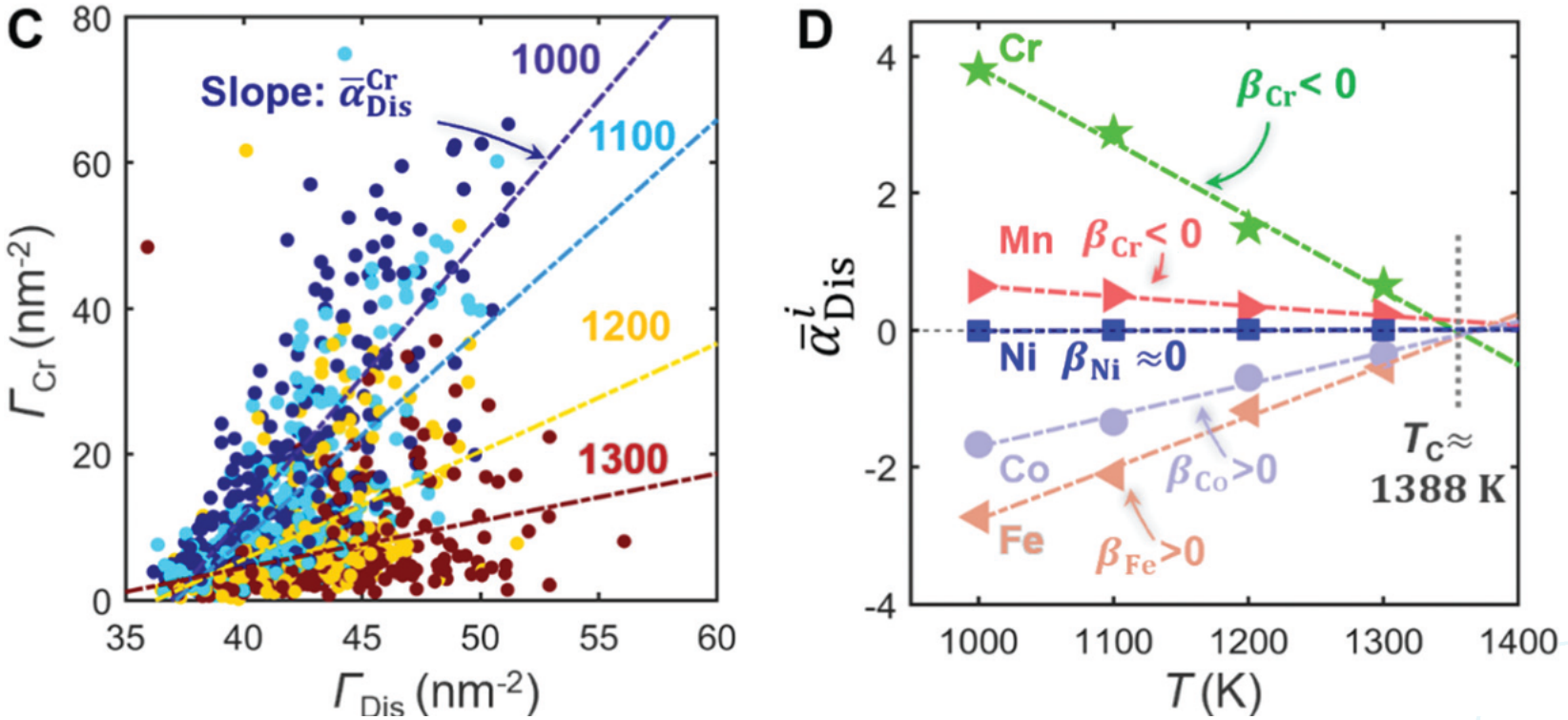






Compensation Effect of GB Segregation in HEAs

- By fitting all 1032 data points, the slope ($\bar{a}_{\mathrm{Dis}}^{\mathrm{Cr}}$) of Γ_{Cr} vs Γ_{Dis} decreases as a function of temperature.
- $\bar{a}_{\mathrm{Dis}}^{\mathrm{Cr}}$ and $\bar{a}_{\mathrm{Dis}}^{\mathrm{Mn}}$ decrease with increasing temperature, $\bar{a}_{\mathrm{Dis}}^{\mathrm{Fe}}$ and $\bar{a}_{\mathrm{Dis}}^{\mathrm{Co}}$ increase with increasing temperature
- If we calculate $\bar{a}_{\mathrm{Dis}}^{\mathrm{Cr}}$ values and plot it as a function of temperature for all five elements, five lines cross over at a same point, where temperature is around 1388 K (called compensation temperature T_c)



Physics-Informed Data-Driven Model (PIDDM)

PIDDM model expression:

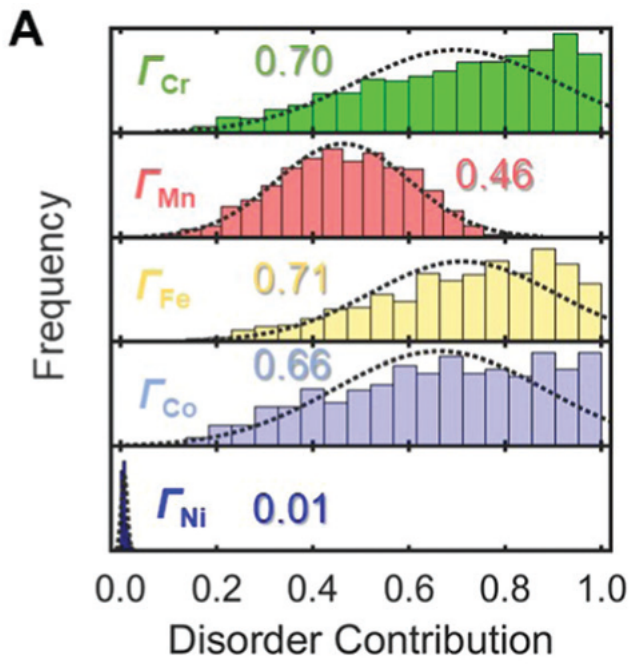
Disorder contribution Compositional contribution

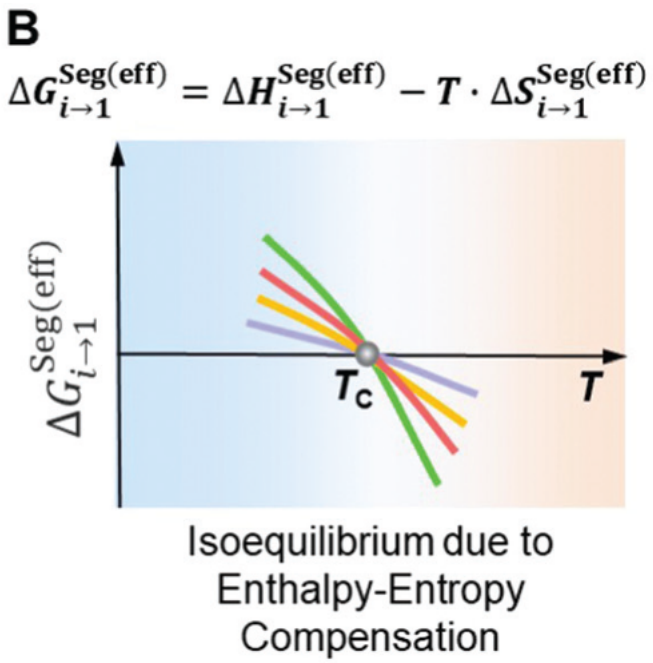
$$\Gamma_i(T,X) = \beta_i \cdot (T - T_C) \cdot \left[\sum_i (\kappa_i^{\mathrm{Dis}} \cdot X_i) \exp\left(-\frac{E_A^{Dis}}{k_B T}\right) - \Gamma_{\mathrm{Dis}}^0\right] + \sum_j \left(\kappa_{i,j}^{\mathrm{Seg}} \cdot X_j\right)$$

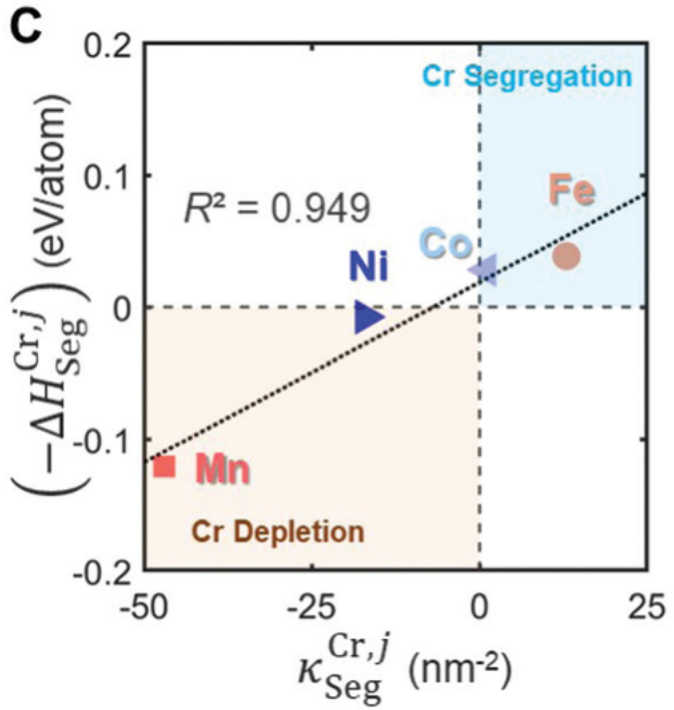
$i =$	T_C (K)	β_i (K^{-1})	$\kappa_{i,Cr}^{Seg}$	$\kappa_{i,Mn}^{Seg}$	$\kappa_{i,Fe}^{Seg}$	$\kappa_{i,Co}^{Seg}$	$\kappa_{i,Ni}^{Seg}$	PIDDM Prediction RMSE (nm^{-2})	ANN Prediction RMSE (nm^{-2})
			($atom/nm^2$)						
Cr	1347	-0.0109	32	-47	13	1	-17	7.4	3.0
Mn	1464	-0.0014	1	-5	8	3	1	1.2	0.6
Fe	1370	+0.0075	-13	26	-26	14	14	5.3	1.9
Co	1371	+0.0046	-15	27	4	-17	12	3.8	1.6
Ni	/	~0	-6.4	-3.7	1.2	-0.5	-7.1	0.88	0.7

Physics-Informed Data-Driven Model (PIDDM)

- Based on PIDDM model, we can predict disorder contribution to GB segregation (see Fig. A below), which around 70% for Cr, Fe, and Co.
- The compensation temperature can be ascribed to enthalpy-entropy compensation



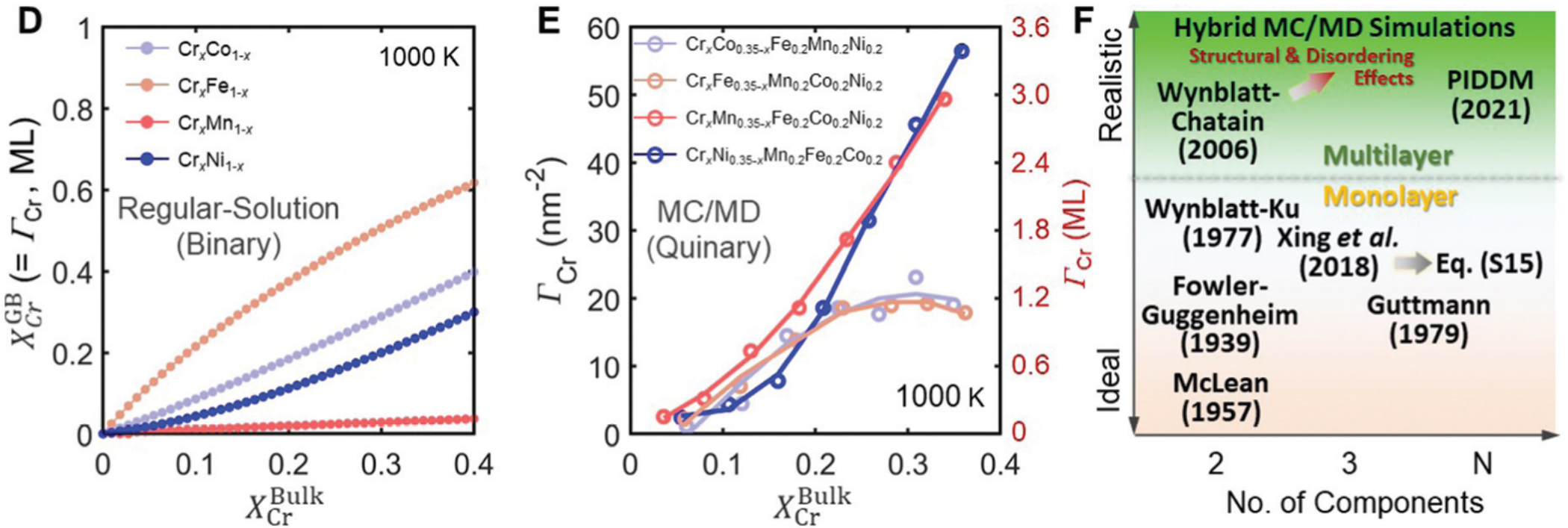




Comparison of PIDDM model vs. Other Segregation Models

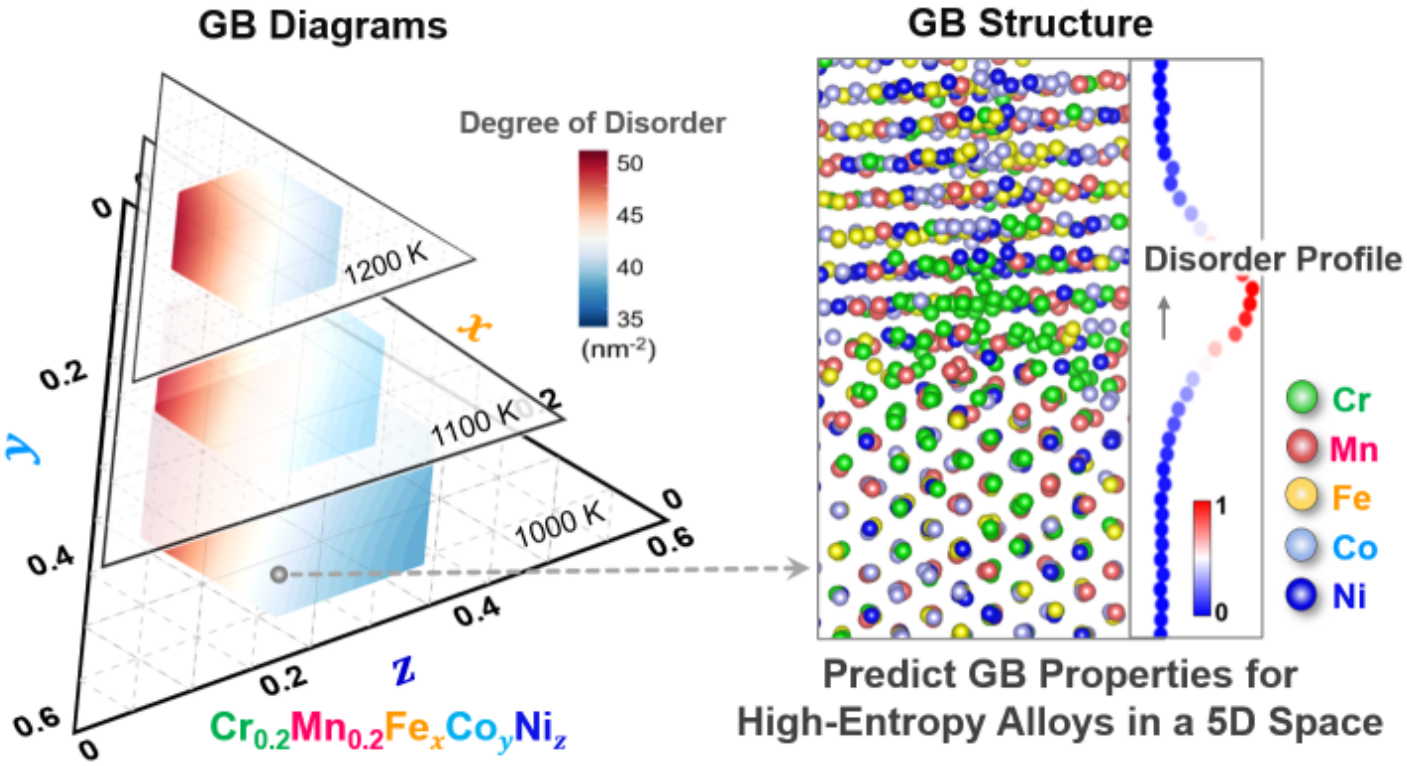
- If calculate Cr segregation as a function of Cr bulk composition in 1000 K based on regular solution model, the segregation trends: Cr_xFe_{1-x} > Cr_xCo_{1-x} > Cr_xNi_{1-x} > Cr_xMn_{1-x}

- Compared to MC/MD simulations, segregation trends are almost reversed. This means regular solution model is not sufficient to predict segregation in HEAs,



Conclusion

- We construct a data-driven framework to predict HEA GB properties as a function of 4 composition and temperature in 5-D space
- ANN is the most accurate model to predict GB properties
- A novel PIDDM has been developed. Compared to ML model, it has physical meaning and can be potentially used for other systems
- Some new physics of GB segregation in HEA have been discovered



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