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Sandia National Laboratories

# Controlling Hydrogen Cottrell Atmospheres Around Dislocations in Austenitic Stainless Steels Through Alloying Using a Combined MD-DFT Pipeline

H-Mat℠ Hydrogen Materials Compatibility Consortium

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**Session:** DS01: Integrating Machine Learning and Simulations for Materials Modeling, Design and Manufacturing

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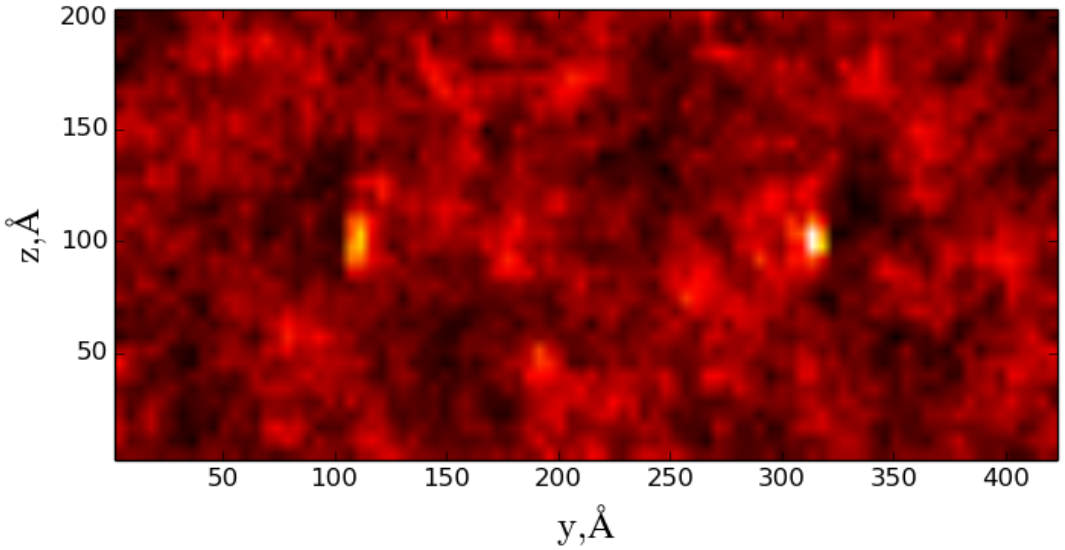
# Hydrogen + Dislocations: Mediating Their Interaction?

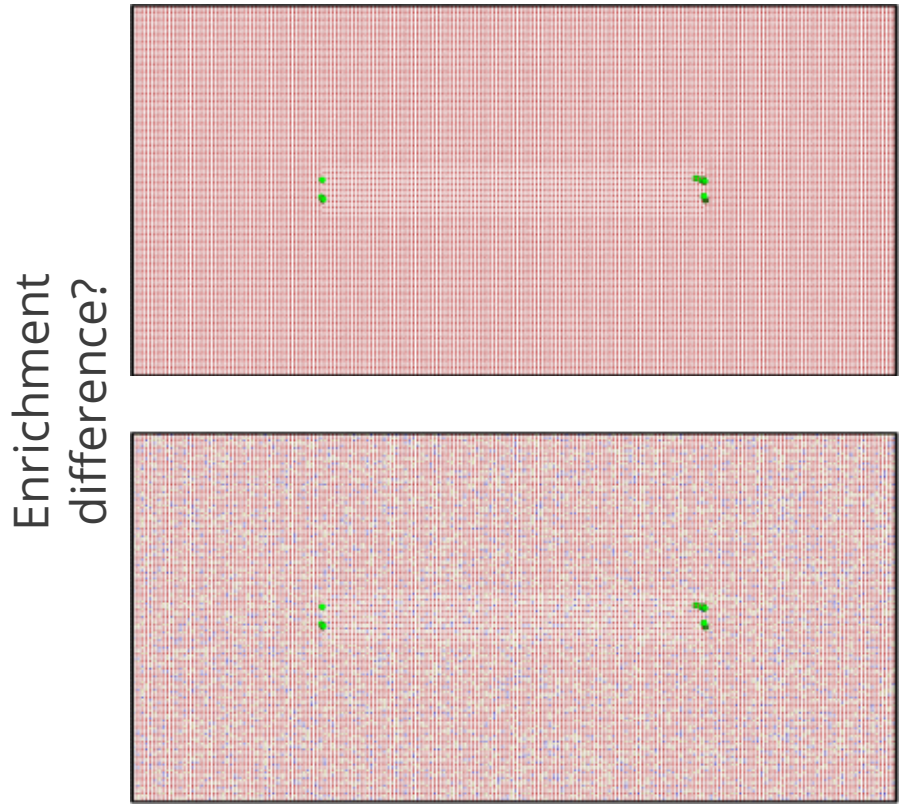
Hydrogen Embrittlement (HE) of cost effective materials hinders proliferation of hydrogen into the energy portfolio

To understand HE we must understand where H is going. Even with low H solubilities, enrichment near defects can result in HE



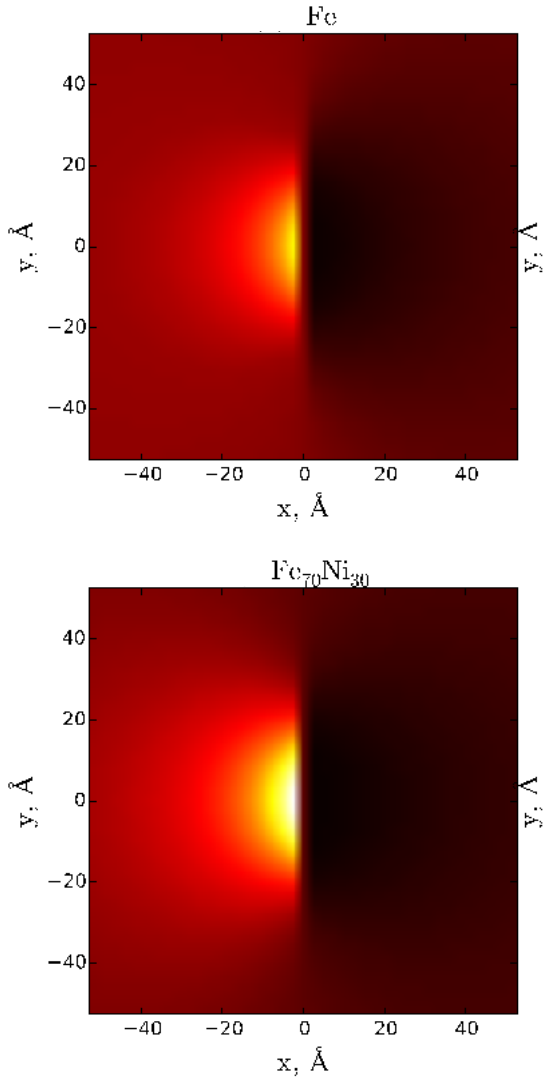

Alloying is commonly used to pull out desirable properties

leverage alloy design to modulate the enrichment near the dislocation core?



# Outline

- MD simulation setup
- Formation of H atmosphere in alloys
- Atmosphere formation affected by alloying?
- Proposal of a machine guided alloy design pipeline

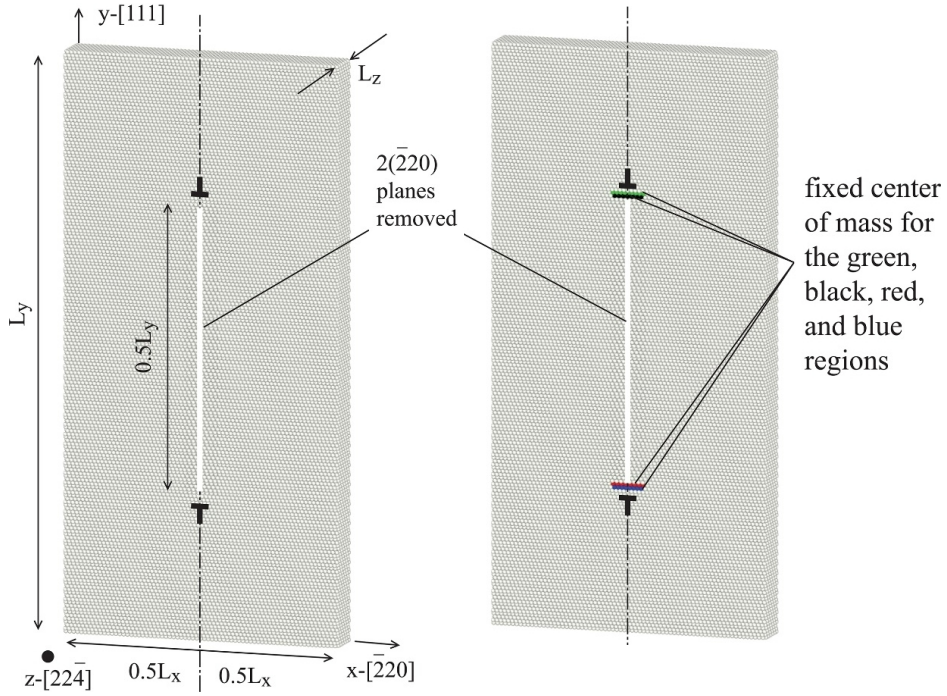



Manuscript for the presented work has been submitted

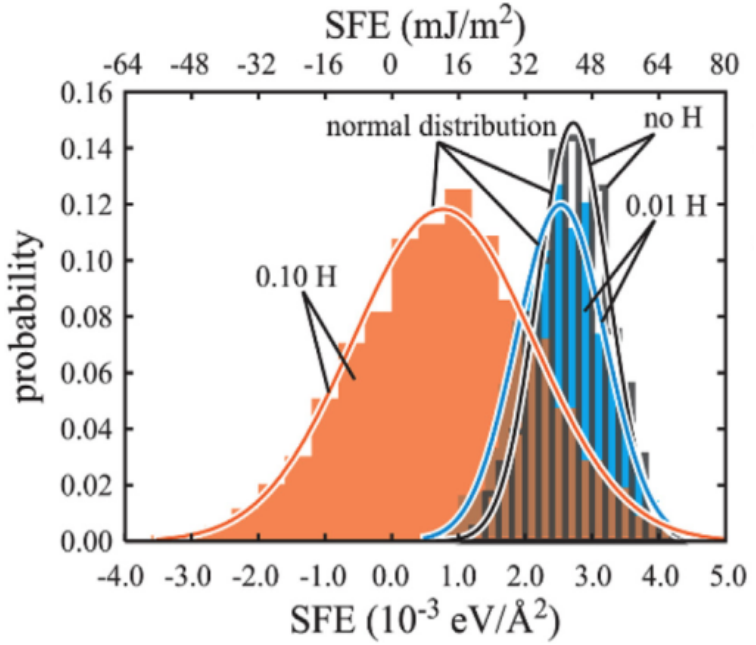
# Simulating Atmospheres

4 main alloy systems:
- Fe: Control system
- $Fe_{70}Ni_{30}$: Just add Ni
- $Fe_{70}Cr_{30}$: Just. Add Cr
- $Fe_{70}Cr_{20}Ni_{10}$: Traditional 304L comp

T=900K
xH=0.01

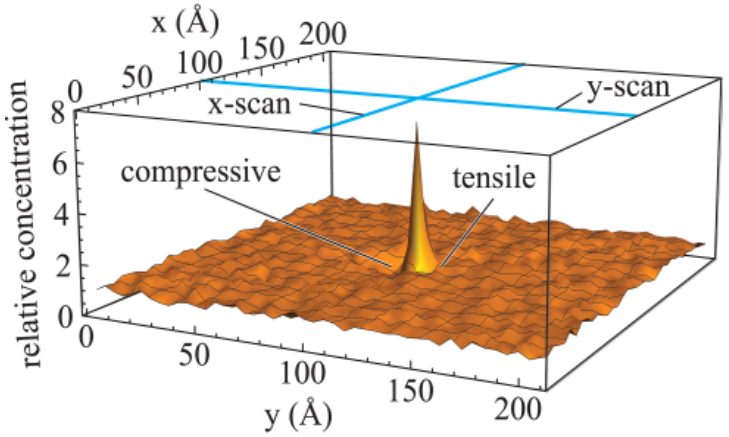



SFE varies strongly with local composition




Due to the local variation in composition a large number of replicas is required

Use established MD methods to avg atmo over 10ns sims, 100 reps



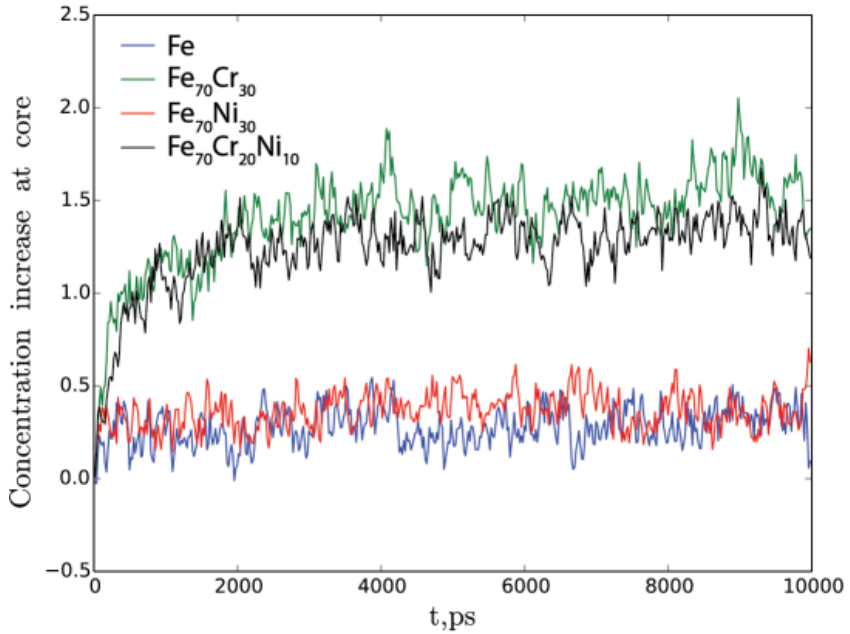

Zhou, et. al. Phys. Chem. Chem. Phys. 2021

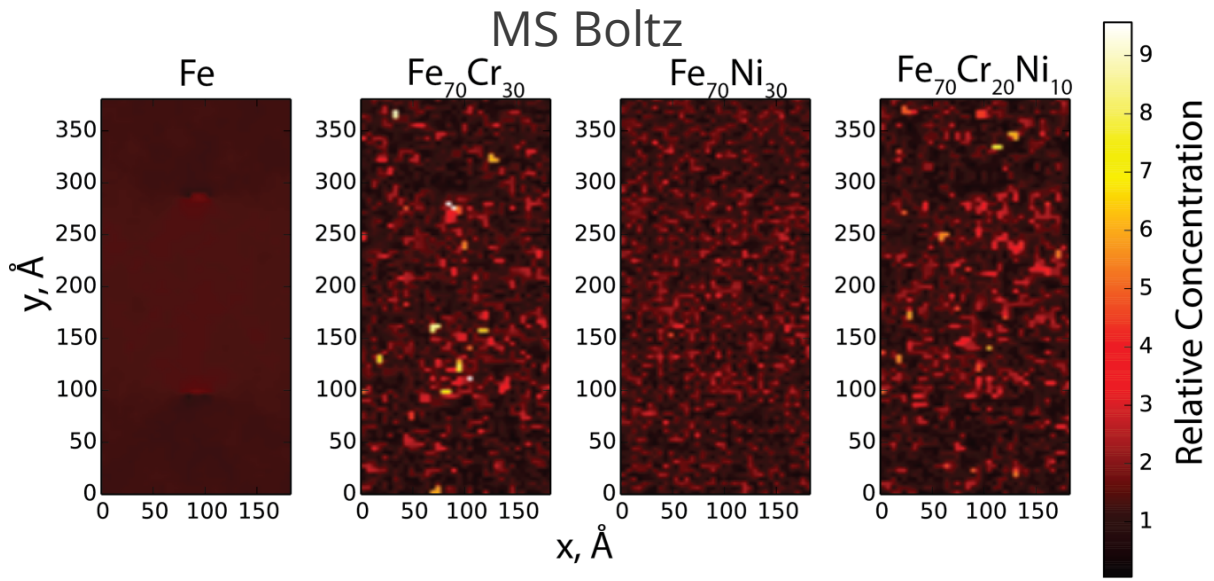
# Alloying Has a Large Effect on Atmosphere

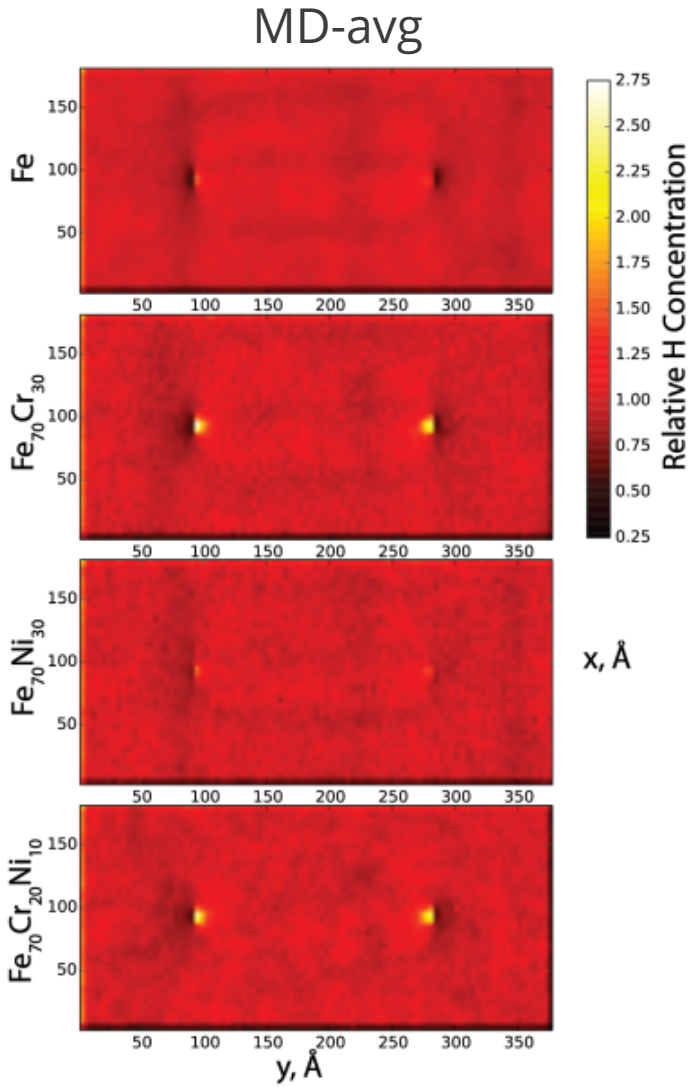
Fe, and $Fe_{70}Ni_{30}$ show much weaker atmosphere

Almost complete disruption of atmo formation seen

Using Molecular Statics doesn't reproduce the same atmosphere



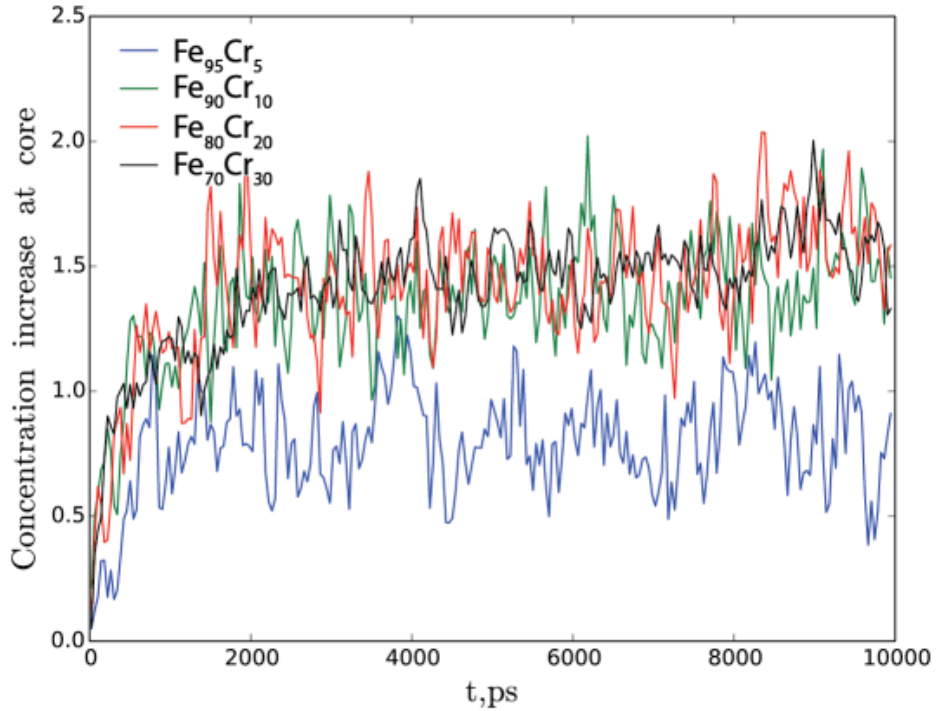



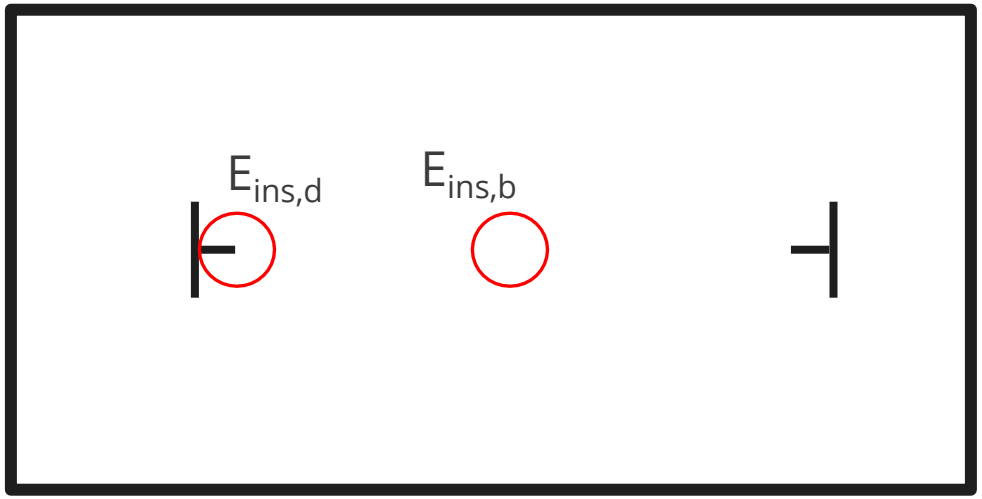
# The effect of Added Cr

Adding Cr changes enrichment, but by how much?

The effect of Cr composition has a saturating effect when xCr>=0.1

Saturation indicates that this is not strictly a linear combination of compositions



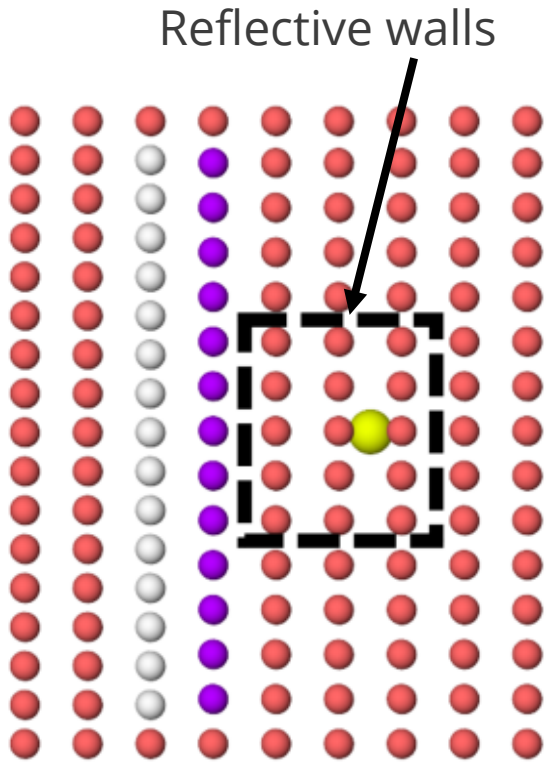




Can we calculate the driving force despite local composition variation?

# MD averaged $E_{ins}$

To measure $E_{ins}$, use reflective walls with many replicas:

- Run with single H in box, avg E
- Delete H, run without H
- E(H=1)-E(H=0)=$E_{ins}$
- Repeat near and far from dislo




The calculated Values of $E_{ins}$ align with the results observed from MD

| | $E_{ins,b}$ (eV) | Error (eV) | $E_{ins,d}$ (eV) | Error (eV) | $E_{ins,d}$-$E_{ins,b}$ (eV) | Error (eV) |
|---|---|---|---|---|---|---|
| Fe | -2.863 | 0.020 | -2.877 | 0.024 | -0.014 | 0.031 |
| $Fe_{70}Ni_{30}$ | -2.801 | 0.020 | -2.829 | 0.029 | -0.028 | 0.035 |
| $Fe_{70}Cr_{30}$ | -2.805 | 0.022 | -2.913 | 0.033 | -0.108 | 0.039 |
| $Fe_{70}Cr_{20}Ni_{10}$ | -2.764 | 0.021 | -2.879 | 0.035 | -0.115 | 0.041 |

Driving Force for non-Cr systems much lower

The question remains:
Why is there a difference?

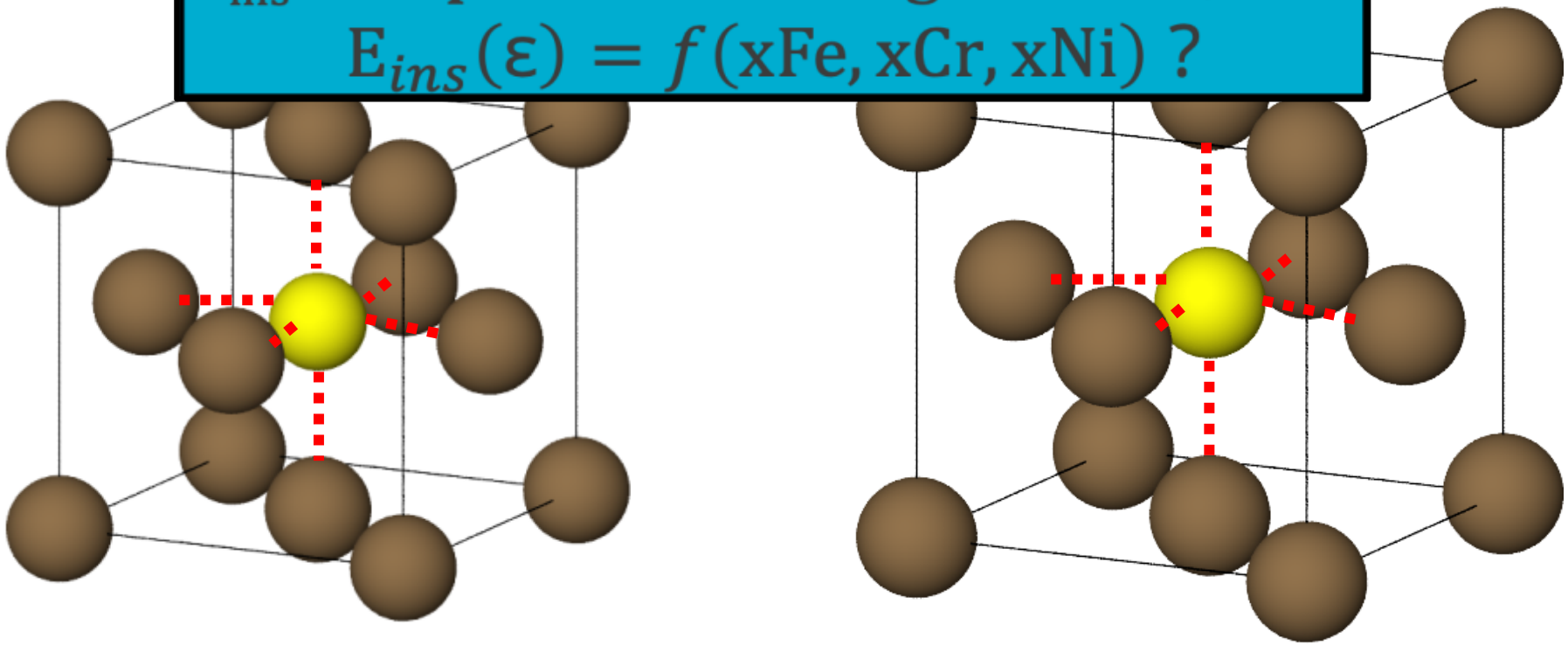
# What's Different?

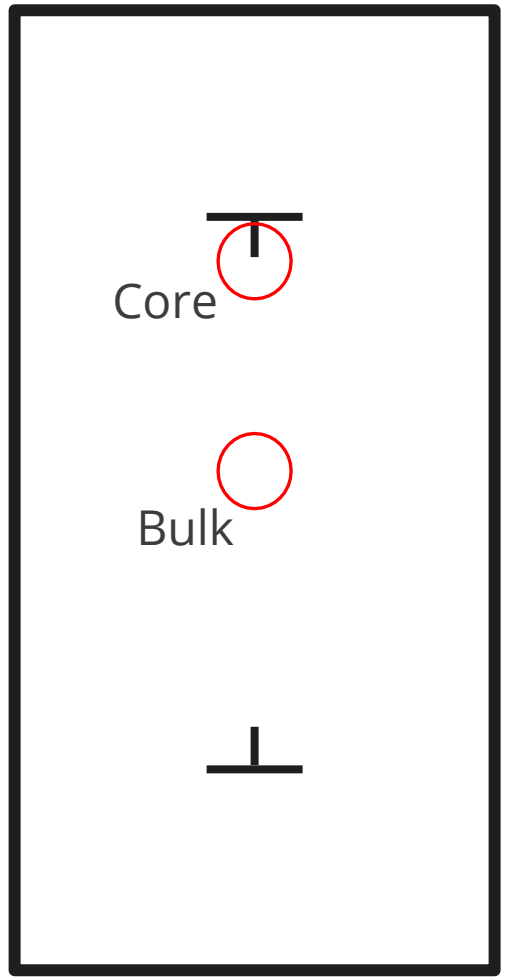
The difference in $E_{ins}$ between the bulk and near the core as a function of alloying is unexpected

The only difference between bulk and the core is a change in local strain.

$E_{ins}$ is ecpected to change with ε , but

$$E_{ins}(\varepsilon) = f(\mathrm{xFe}, \mathrm{xCr}, \mathrm{xNi})\ ?$$



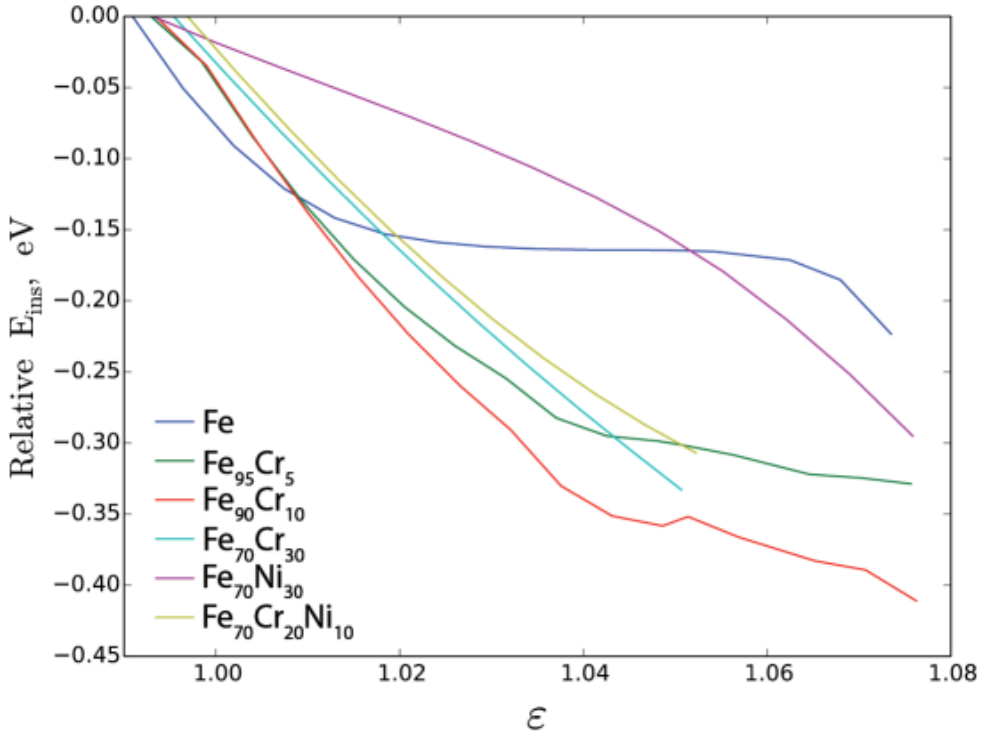


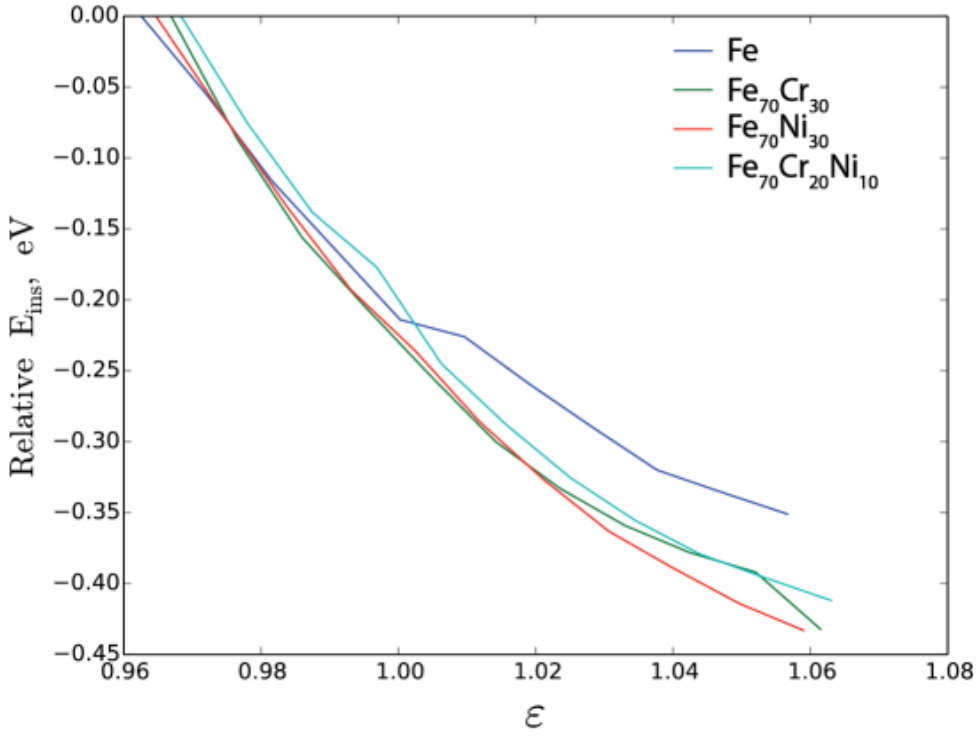
# $E_{ins}$ as a function of ε

Calculate $E_{ins}$ as a function of strain as a function ε using MS and DFT

DFT also predicts that Fe should have a weaker atmosphere:

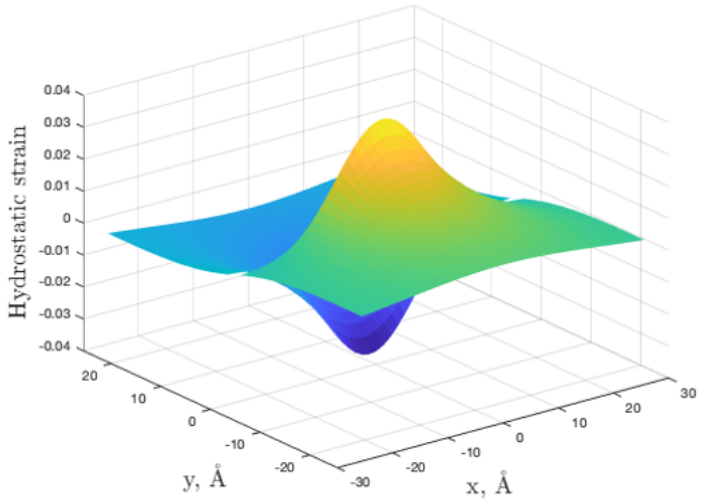
$$\left(\frac{dE_{ins}}{d\varepsilon}\right)_{Fe} < \left(\frac{dE_{ins}}{d\varepsilon}\right)_{304L} \rightarrow \Delta E_{ins,Fe} < \Delta E_{ins,304L} \rightarrow \Delta X_{Fe} < \Delta X_{304L}$$



# Theoretical atmospheres from DFT

Use hydrostatic strain field of a dislocation with $E_{ins}=f(\varepsilon)$:

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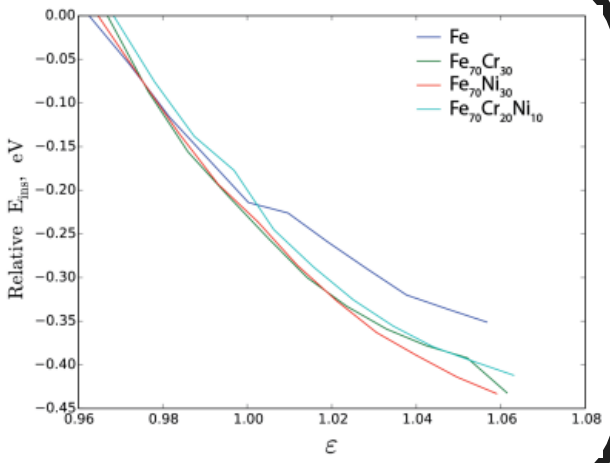
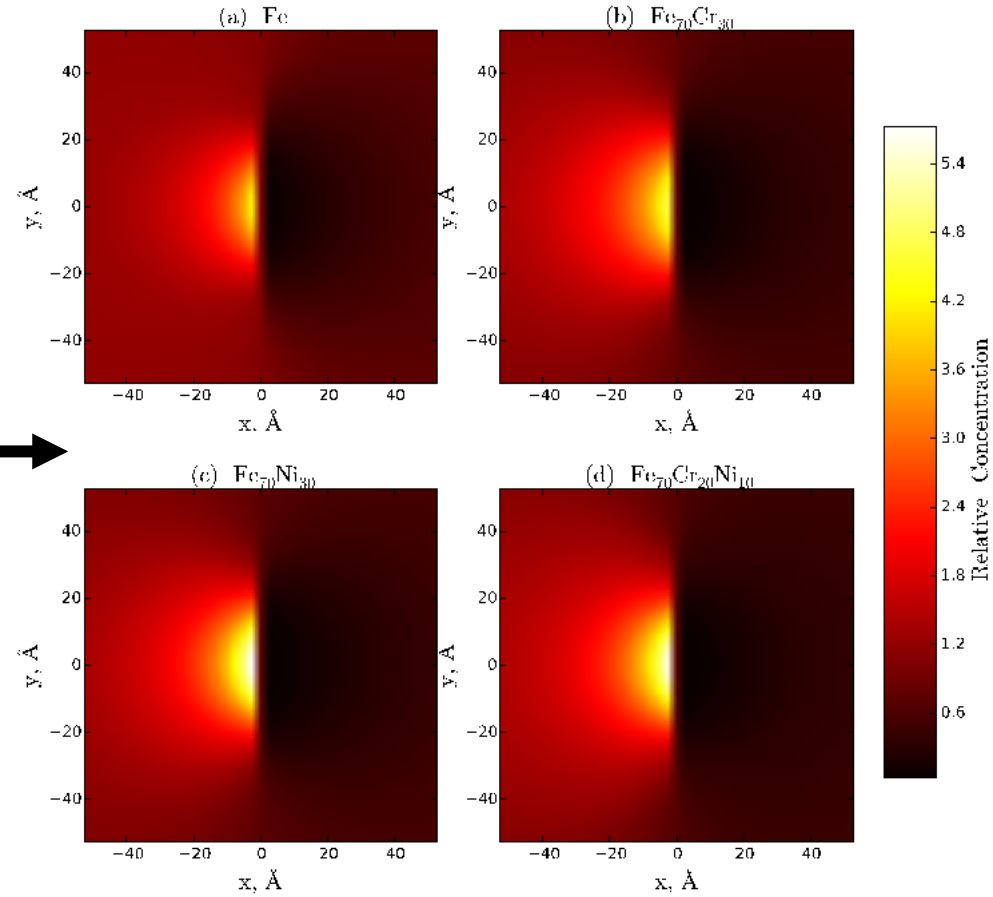



$$X_{H,i} = \frac{\exp\left(-\frac{E_{ins,i}}{k_B T}\right)}{\sum_n \exp\left(-\frac{E_{ins,n}}{k_B T}\right)}$$




Atmosphere behavior can be predicted from DFT calculations

These atmospheres match well the results from MD

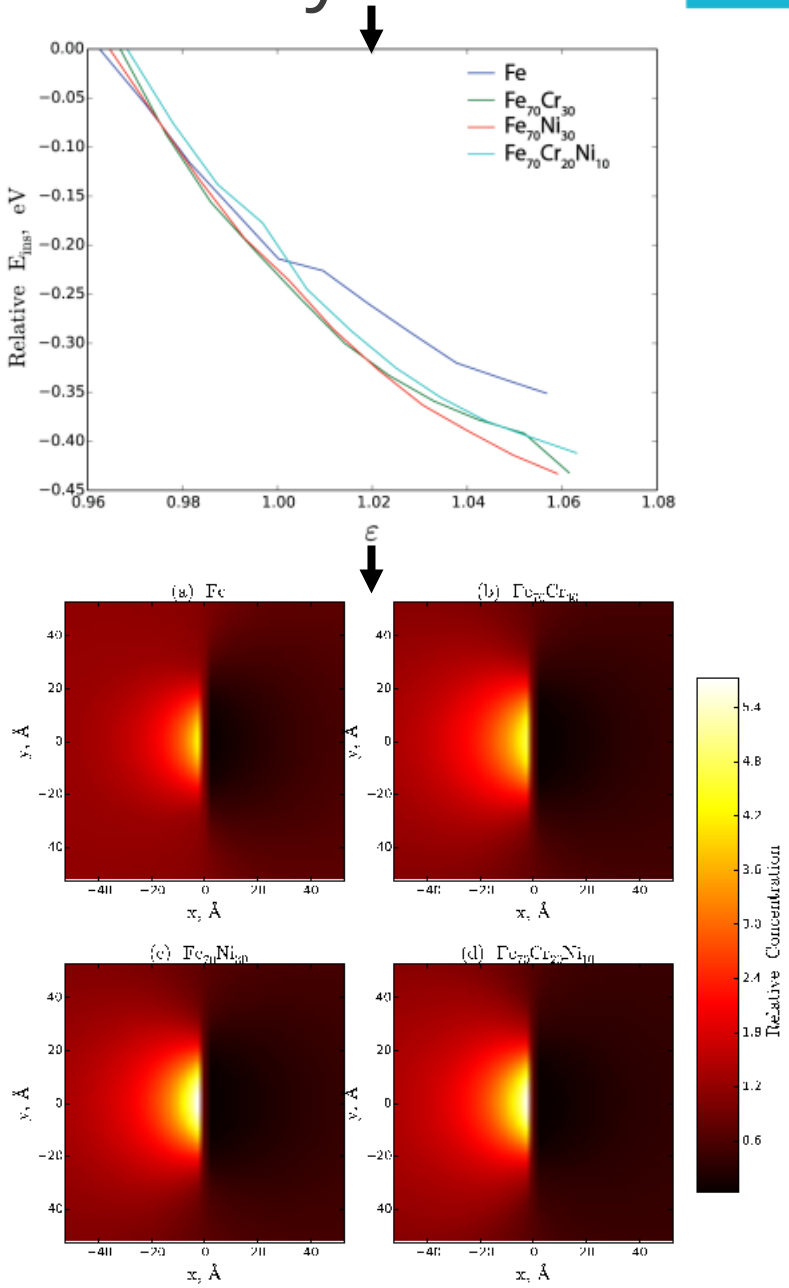
→ Pipeline for estimating atmospheres without an interatomic potential

Varvenne, et. al. Acta Mat 2016

# Conclusions

- Simulated the transient formation of hydrogen Cottrell atmospheres in various alloys
- Alloying can modulate H enrichment at dislo
- Careful MD simulation confirm difference in how $E_{ins}$ changes with alloying
- Simple DFT calculations can be used to predict atmo and spearhead alloy design

Alloy + DFT



# Acknowledgements

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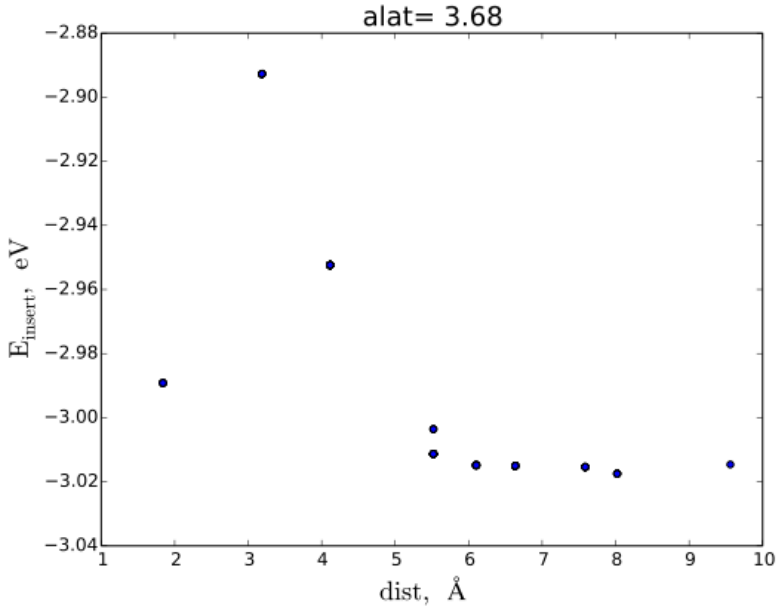
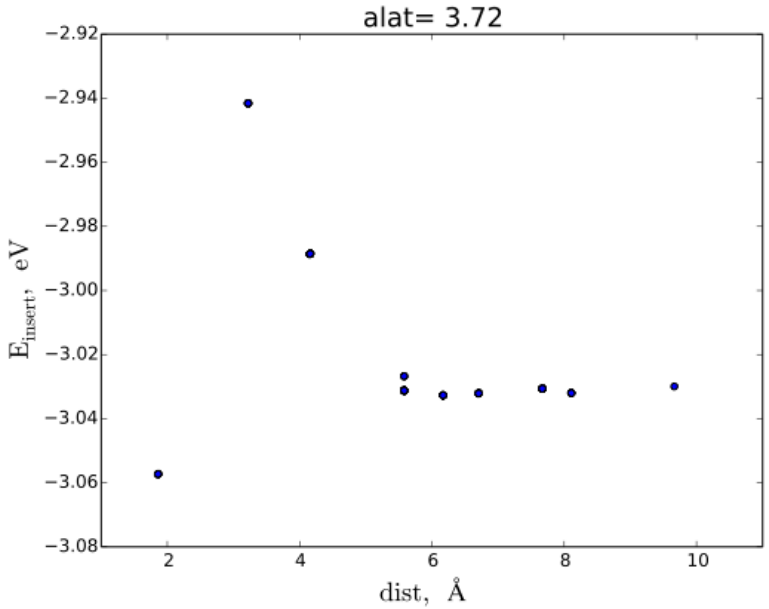
Thank you to DOE's H-MAT program for funding this work.

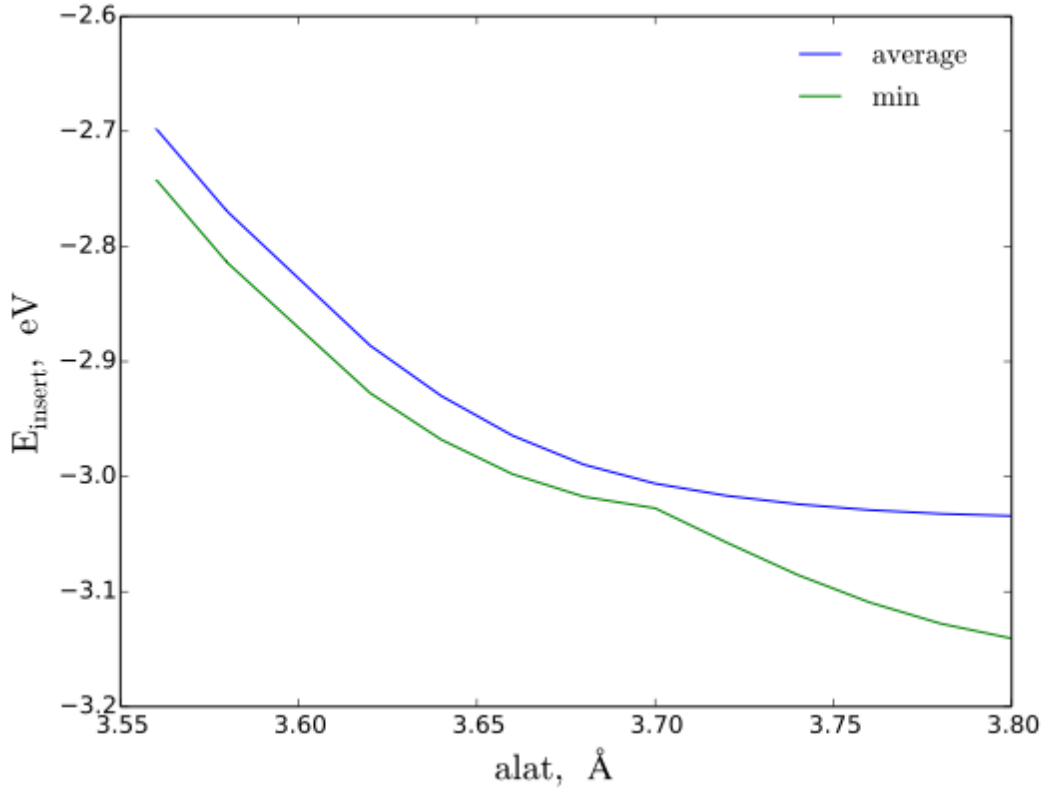
# Thanks for your time, questions?

# Extra Slides

# Microscopic level of what's happening with Cr



# Convergence Calculations

With more replicas the trend becomes apparent

With more replicas the curves smooths out

Alloying will increase the noise