Introduction to ESPEI

Database development and uncertainty quantification

Brandon Bocklund

Materials Genome Foundation

July 13-14, 2022

“Software Tools from Machine Learning to Phase Diagrams” Virtual Workshop

CALPHAD modeling

Derivatives of Gibbs energy
enthalpy, entropy, heat capacity

Phase equilibrium data
phase boundaries

Gibbs Energy of Individual Phases

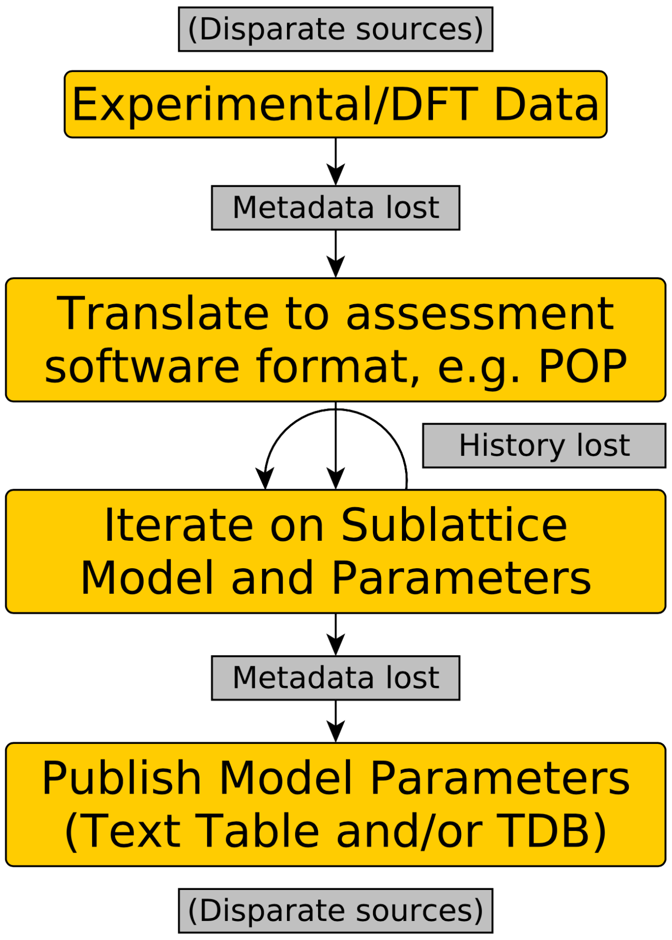
$$G^{\phi}(T,P,x_i) = G^{\mathrm{srf}} + G^{\mathrm{cnf}} + G^{\mathrm{phys}} + G^{\mathrm{xs}}$$

Equilibrium, phase diagrams,
driving forces, physical/chemical properties

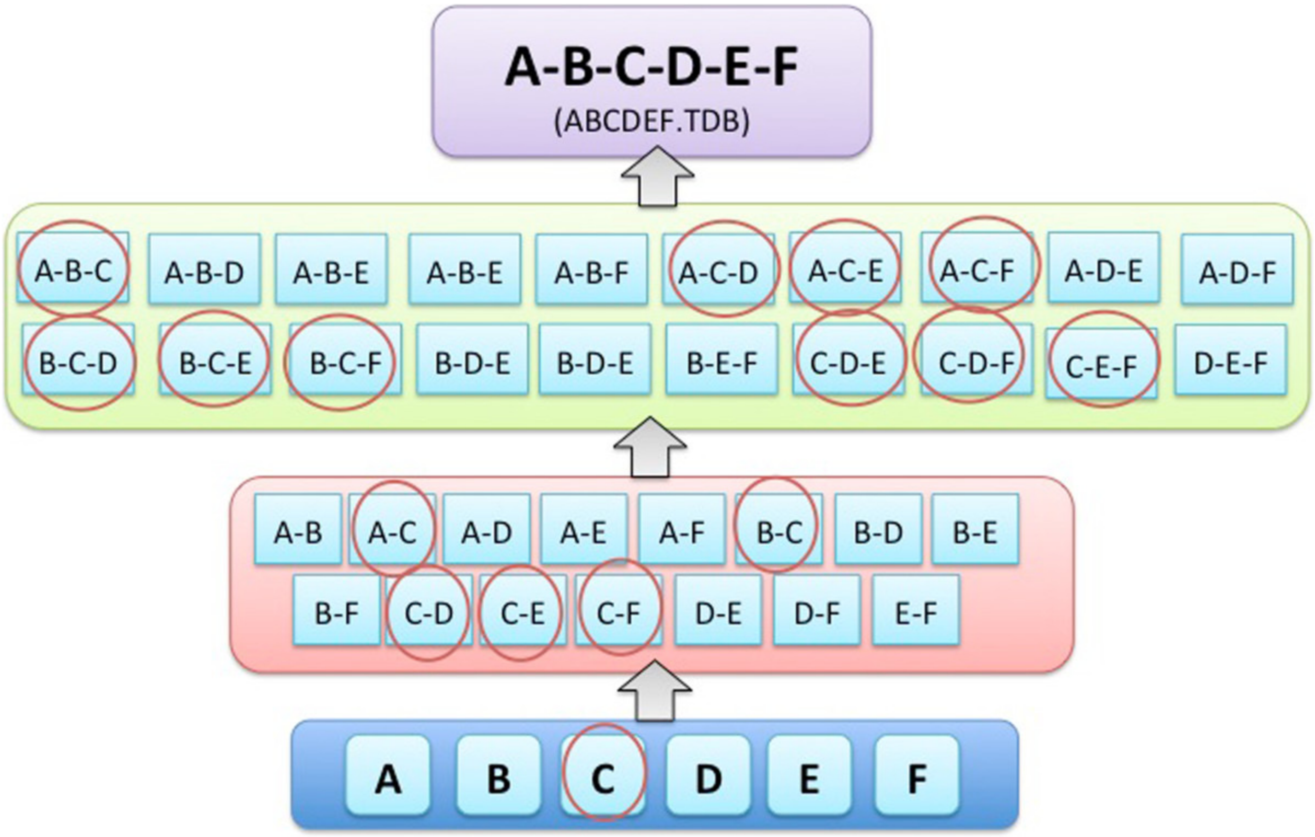
Pure elements → Binary → Ternary → Multicomponent

Kaufman & Bernstein: *Computer Calculation of Phase Diagrams (1970)*

CALPHAD database development process



Otis, PhD Thesis (2016)

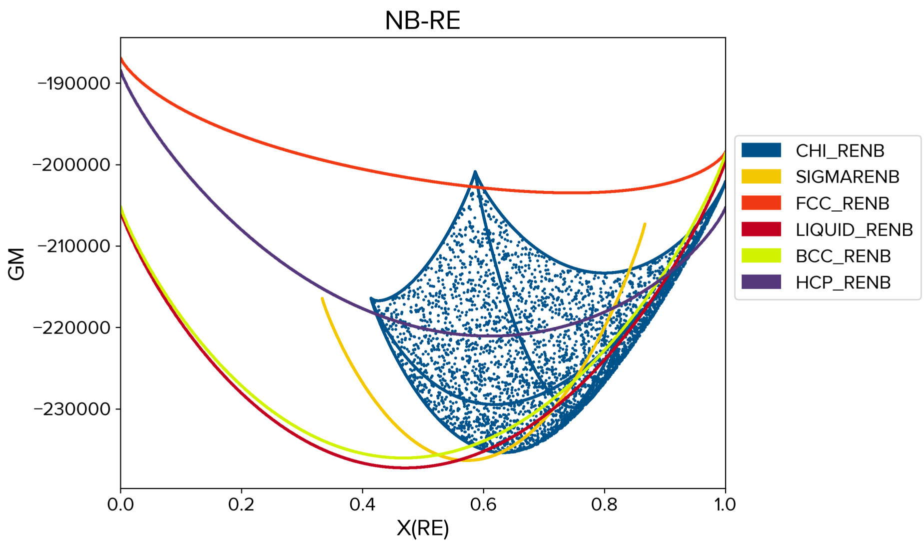


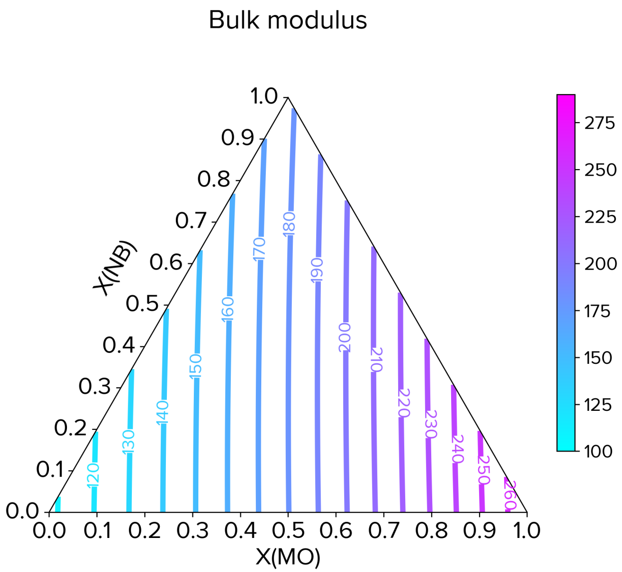
Campbell, IMMI (2014)

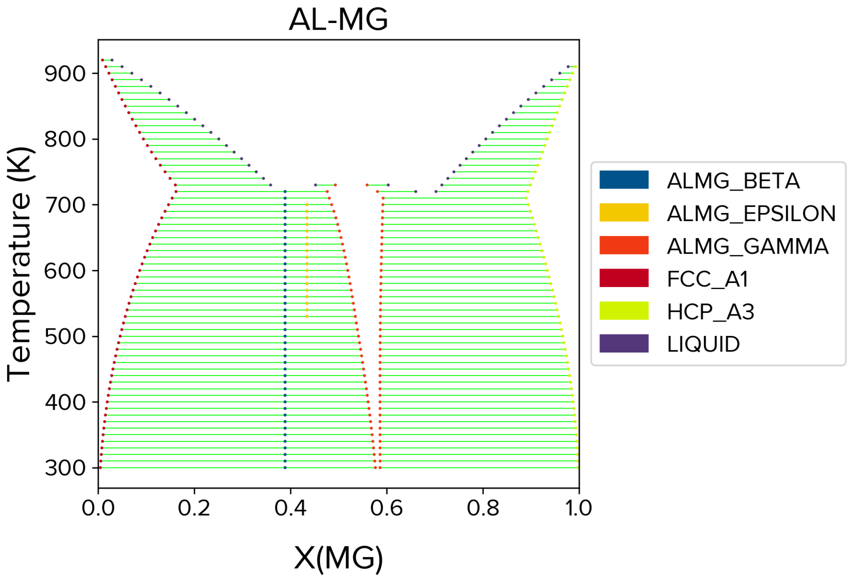
pycalphad: open-source Calphad library

- Initially released in April 2015
- Built on and integrated with the scientific Python stack
- Defines Calphad models symbolically using SymPy
- Fast numerical core written in Cython

pycalphad







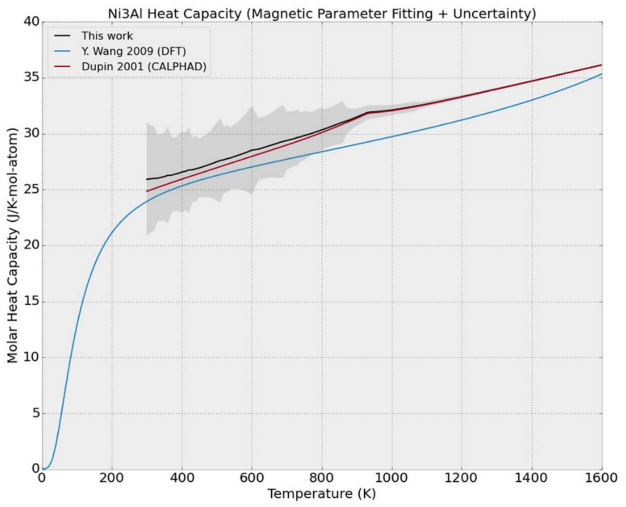
https://pycalphad.org

ESPEI: Database Development and UQ

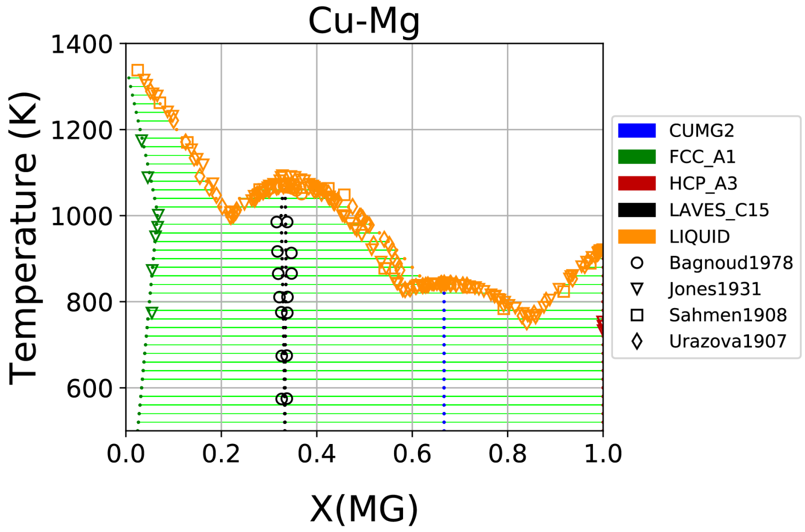
Extensible Self-optimizing Phase Equilibria Infrastructure

1. Parameterize Calphad models from single phase data
2. MCMC: Optimize and quantify uncertainty
3. Propagate uncertainty to properties

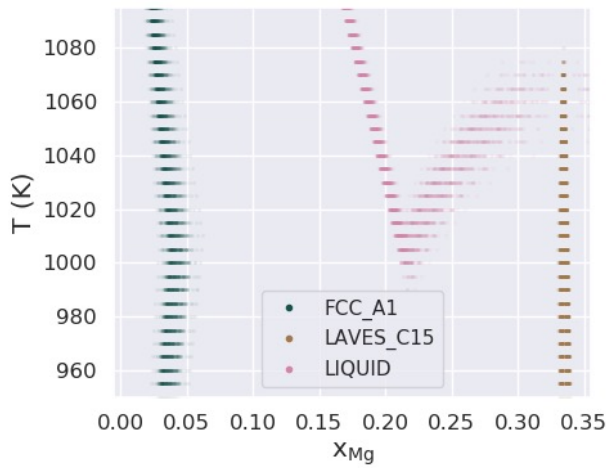
https://espei.org



Otis, Liu, JOM (2017)



Bocklund, MRS Comm (2019)



Paulson, Acta Mat. (2019)

Impact of ESPEI

Journal of MATERIALS RESEARCH

Invited Feature Paper

DOI: 10.1557/s43578-020-00073-6

EARLY CAREER SCHOLARS IN MATERIALS SCIENCE

Sensitivity estimation for calculated phase equilibria

Richard Otis[1,a)], Brandon Bocklund[2], Zi-Kui Liu[2]

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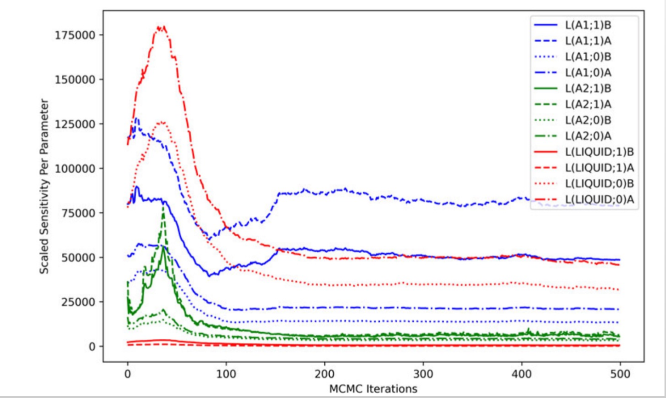
[2]Department of Materials Science and Engineering, Pennsylvania State University, University Park, Pennsylvania 16802, USA



Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jalcom

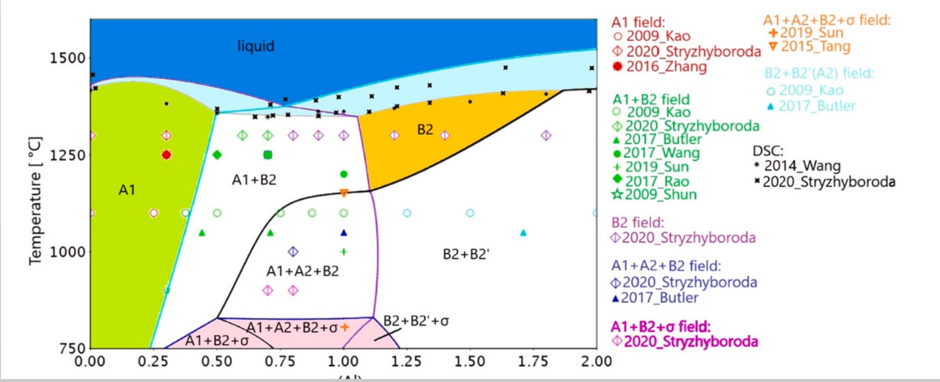
ELSEVIER

Thermodynamic modeling of the Al-Co-Cr-Fe-Ni high entropy alloys supported by key experiments

Marlena Ostrowska[a,*], Paola Riani[a], Brandon Bocklund[b], Zi-Kui Liu[b], Gabriele Cacciamani[a]

[a] Department of Chemistry and Industrial Chemistry, University of Genoa, Italy

[b] Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16802, USA



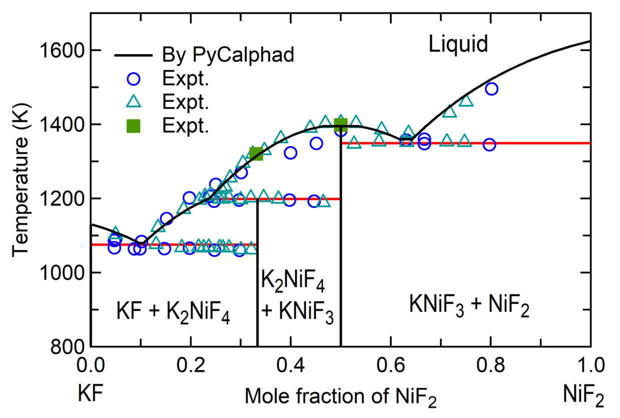
arXiv > physics > arXiv:2204.09111

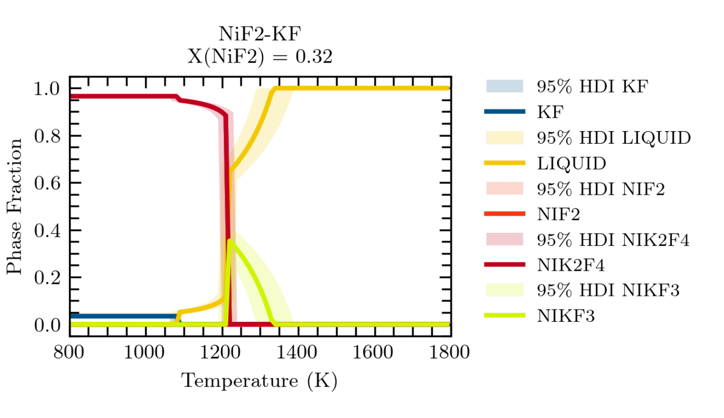
Physics > Computational Physics

[Submitted on 19 Apr 2022]

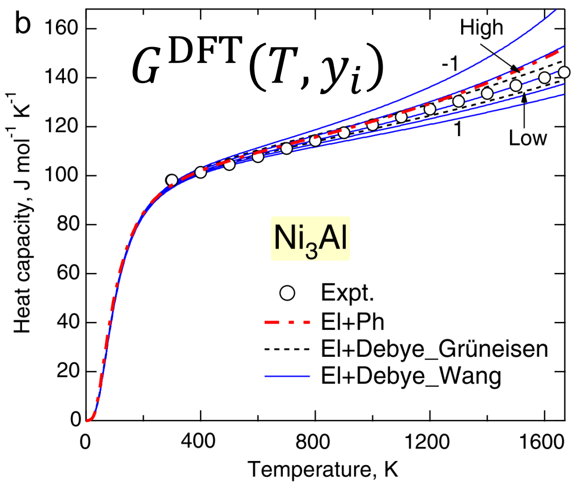
Thermodynamic modeling with uncertainty quantification using the modified quasichemical model in quadruplet approximation: Implementation into PyCalphad and ESPEI

Jorge Paz Soldan Palma, Rushi Gong, Brandon J. Bocklund, Richard Otis, Max Poschmann, Markus Piro, Yi Wang, Tatiana G. Levitskaia, Shenyang Hu, Hojong Kim, Shun-Li Shang, Zi-Kui Liu





Parameter selection in ESPEI



Shang, Comput. Mater. Sci. (2009)

Fit non-equilibrium thermochemical data to a functional form

Temperature dependence: power expansion

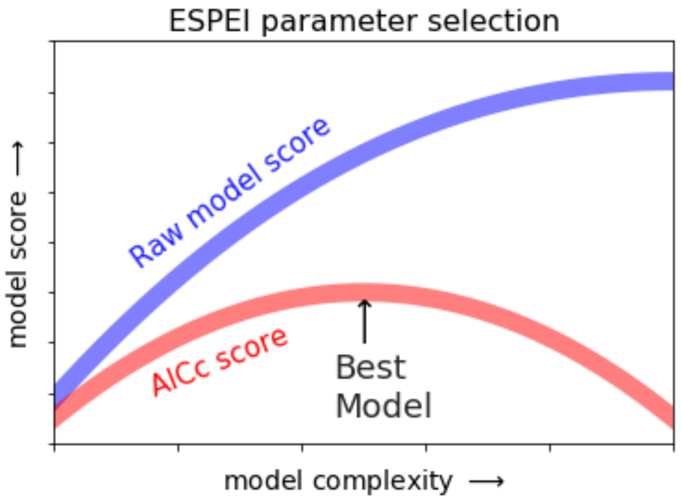
$$G(T) = a + b\,T + c\,T\,\ln T + d\,T^2 + e\,T^{-1}$$

Composition dependence: Redlich-Kister polynomial

$$G^{\mathrm{xs}}(T, y_i) = \sum_{i,j \neq i} y_i y_j \left(y_i - y_j\right)^v \, {}^vL$$

$${}^vL\,(T) = a + b\,T + \ \ldots$$

Overfitting prevented with corrected Akaike information criterion



VanderPlas (2016)

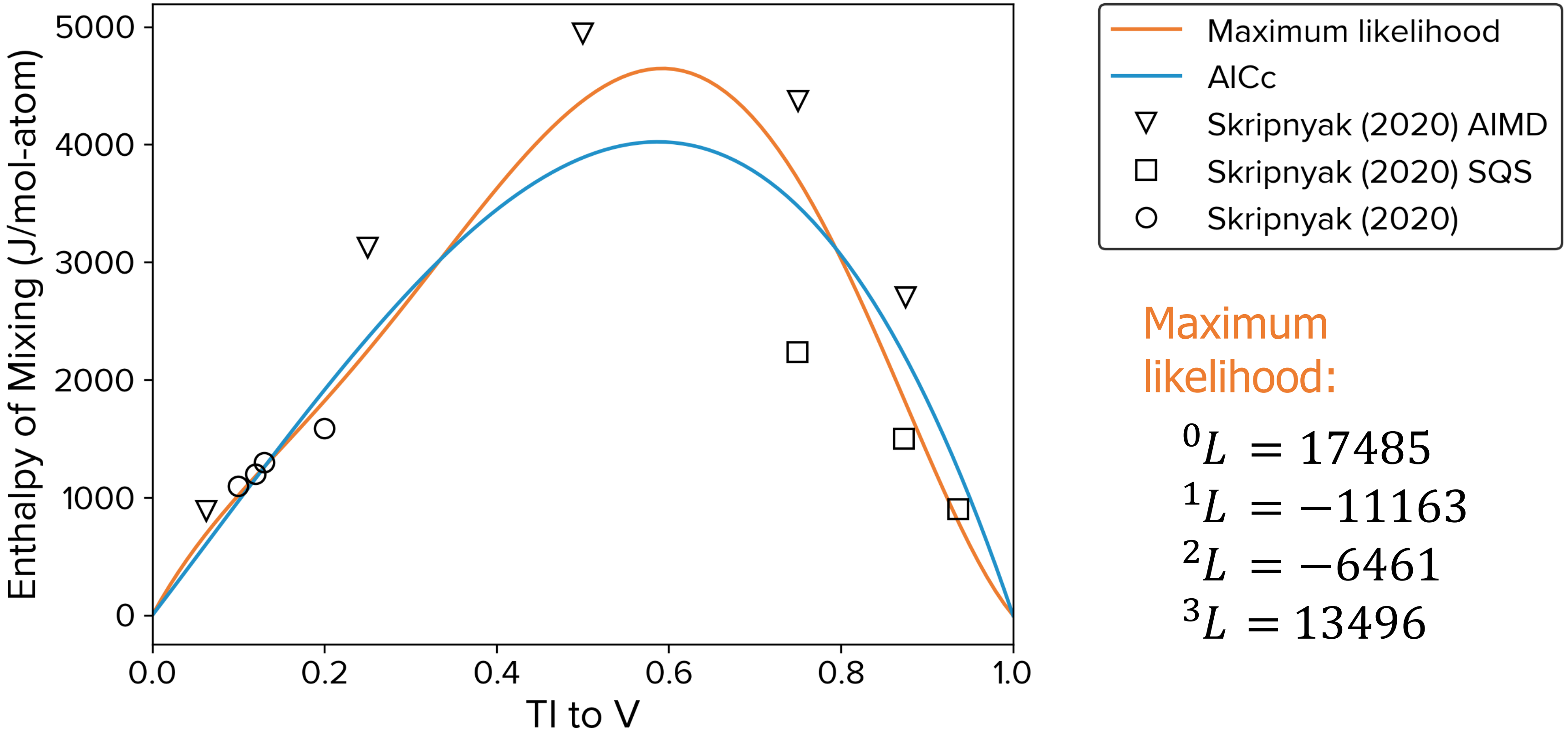
$$\mathrm{AICc} = n \ln \frac{\mathrm{RSS}}{\mathrm{n}} + 2k + \frac{2k^2 + 2k}{n - k - 1}$$

RSS – Sum of square residuals

k – # model parameters (model complexity)

n – # data points

Parameter selection for bcc Ti-V



$$\sum_{i,j\neq i} y_i y_j \left(y_i - y_j\right)^v \, {}^vL$$

Maximum likelihood:

$${}^0L = 17485$$
$${}^1L = -11163$$
$${}^2L = -6461$$
$${}^3L = 13496$$

AICc:

$${}^0L = 15552$$
$${}^1L = -5977$$

Interactive Demo

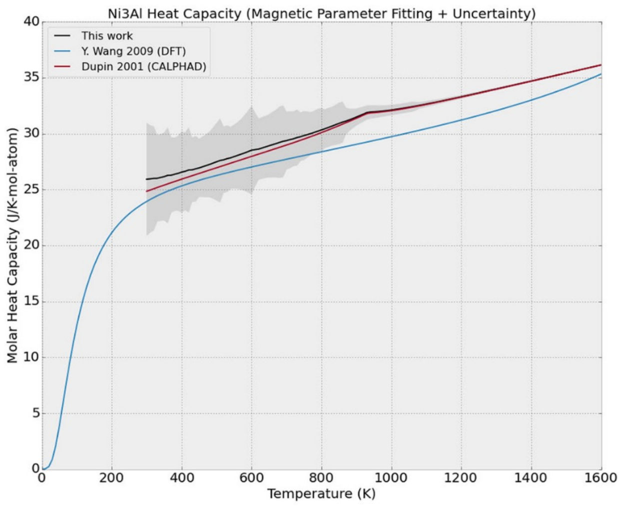
ESPEI Parameter Generation

ESPEI: Database Development and UQ

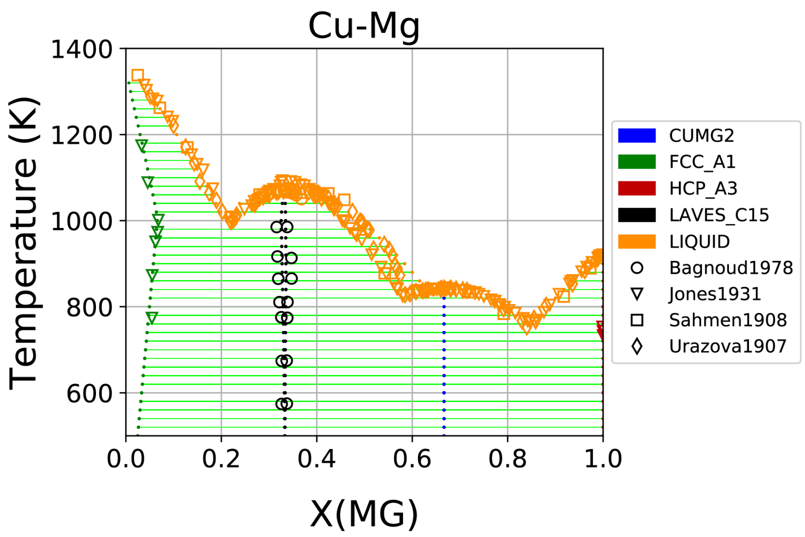
Extensible Self-optimizing Phase Equilibria Infrastructure

1. Parameterize Calphad models from single phase data
2. **MCMC: Optimize and quantify uncertainty**
3. Propagate uncertainty to properties

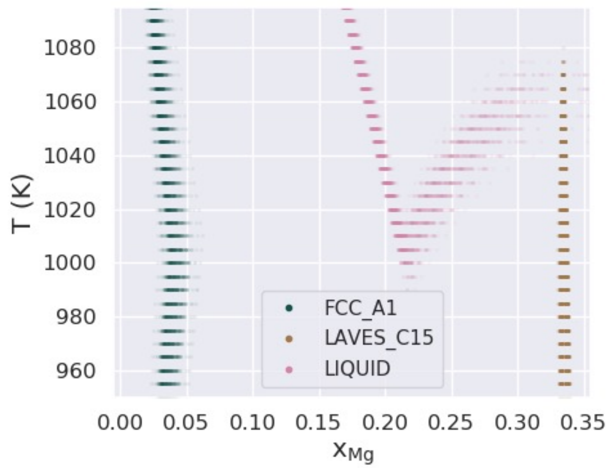
https://espei.org



Otis, Liu, JOM (2017)



Bocklund, MRS Comm (2019)

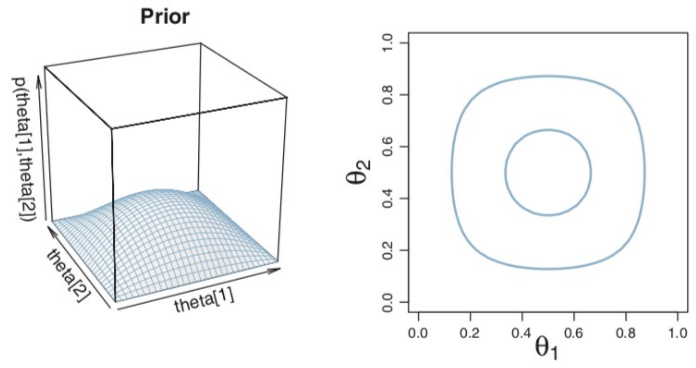


Paulson, Acta Mat. (2019)

MCMC: Bayesian parameter estimation

Posterior | Likelihood | Prior | Data

$$\Pr(\boldsymbol{\theta}|\mathbf{D}) = \frac{\Pr(\mathbf{D}|\boldsymbol{\theta})\ \Pr(\boldsymbol{\theta})}{\Pr(\mathbf{D})}$$

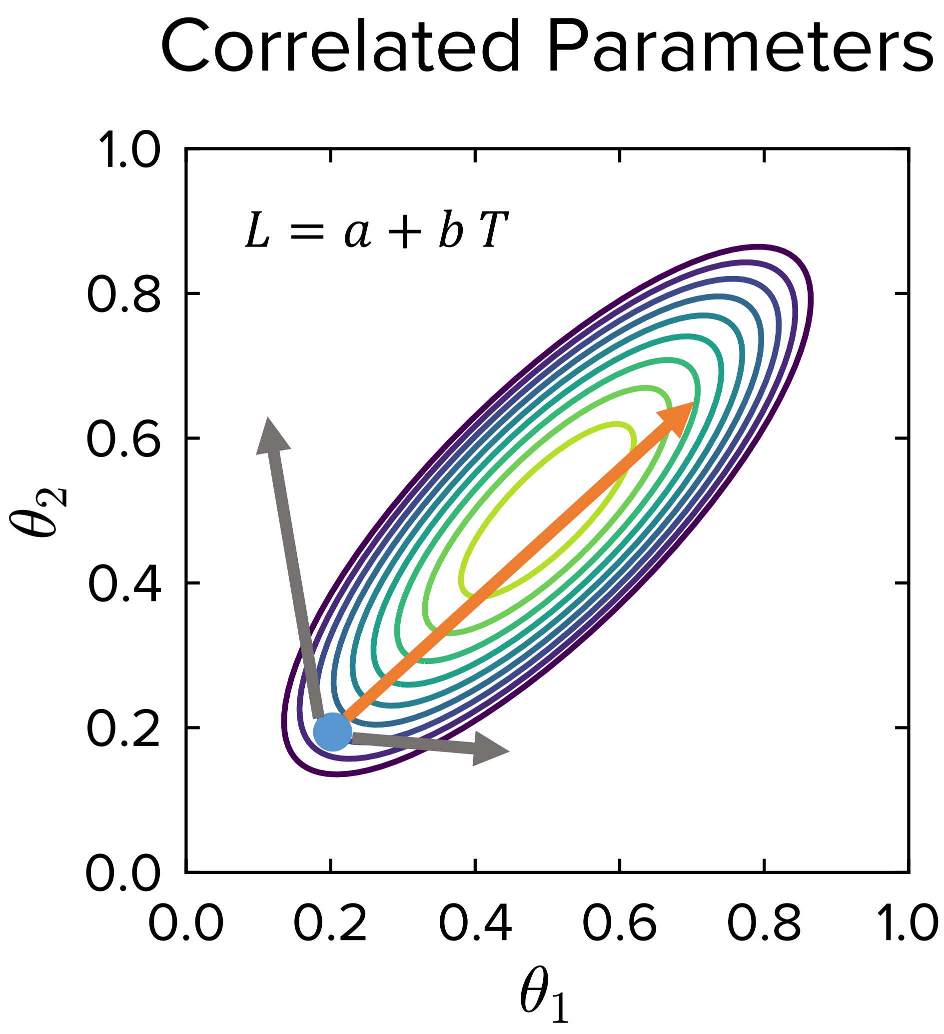


- Markov chain
  - Sequence of values that are independent from each other
- Monte Carlo
  - Random sampling

Core principle: the probability of a parameter value is proportional to the number of times the Markov chains visits that value

Kruschke, Academic Press (2015)

Ensemble MCMC: efficient exploration of correlated parameter space

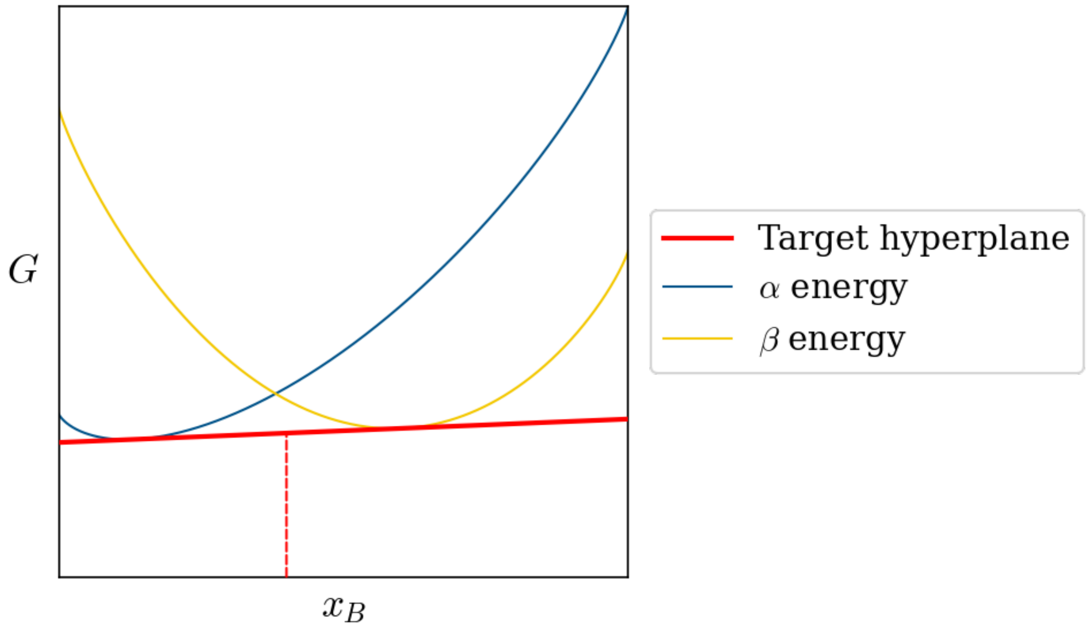


- Use many (an ensemble of) parameter chains simultaneously
  - Proposals are stretched by an affine transformation to a random chain in the ensemble
  - Key property: **affine invariance**
- In ESPEI: the ensemble is generated by multi-variate normal distribution from the initial parameters
  - Mean is at the parameter initial value
  - Standard deviation is a factor of the mean value
  - Wider initial ensemble → better coverage of parameter space, but may be far from the initial local minimum

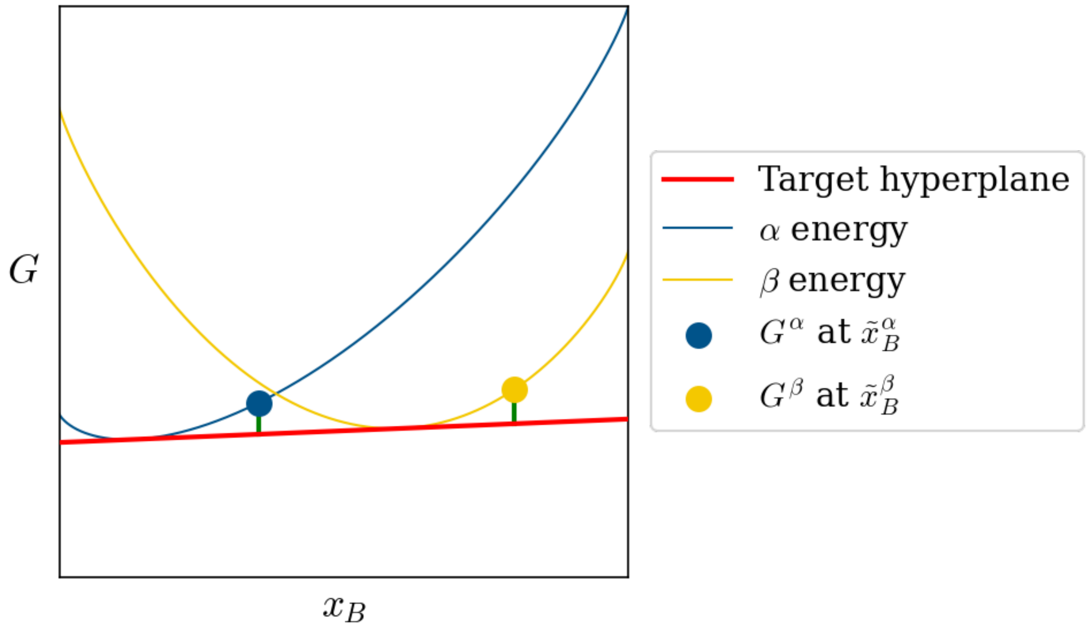
Calculation of phase equilibria error

Goal: Find the parameters that minimize the driving forces to the target hyperplane defined by the experimental phase compositions

Target hyperplane: $\tilde{x}$ known

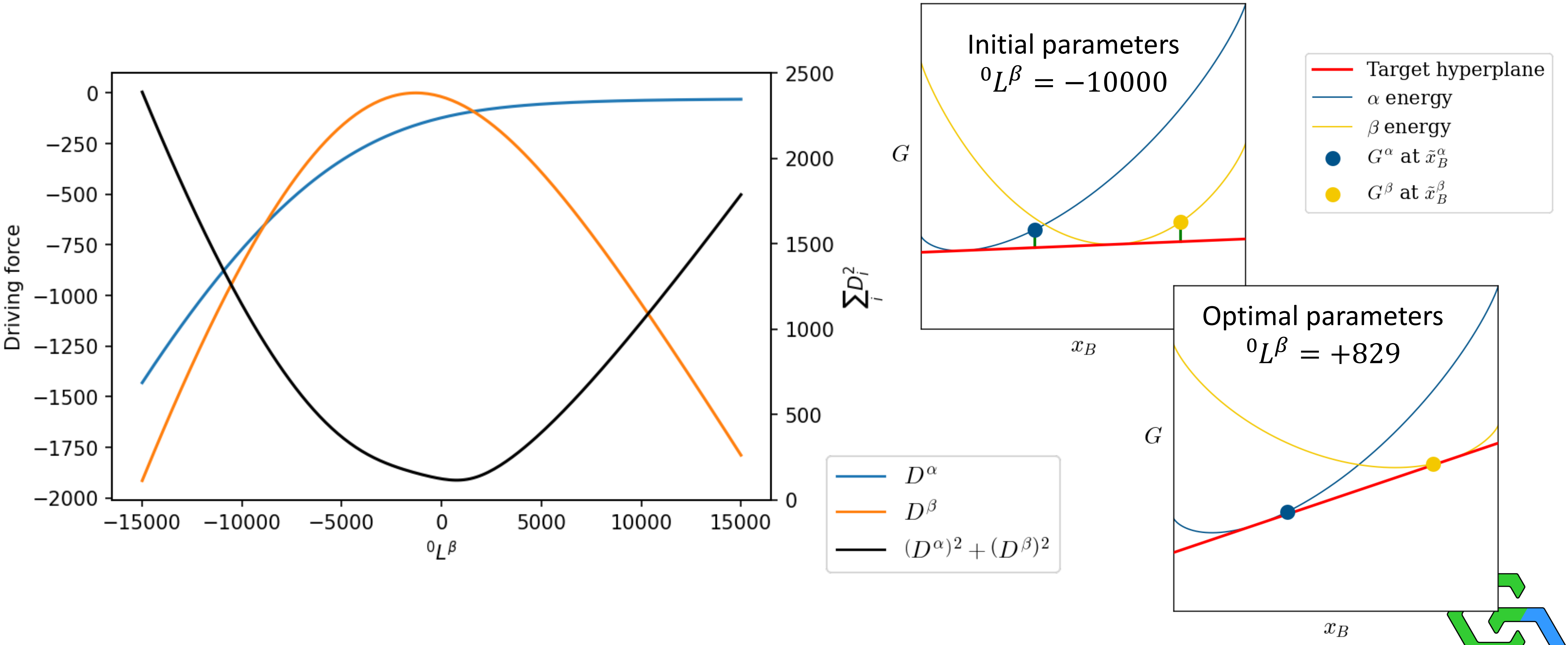


Driving forces: $\tilde{x}^{\phi}$ known



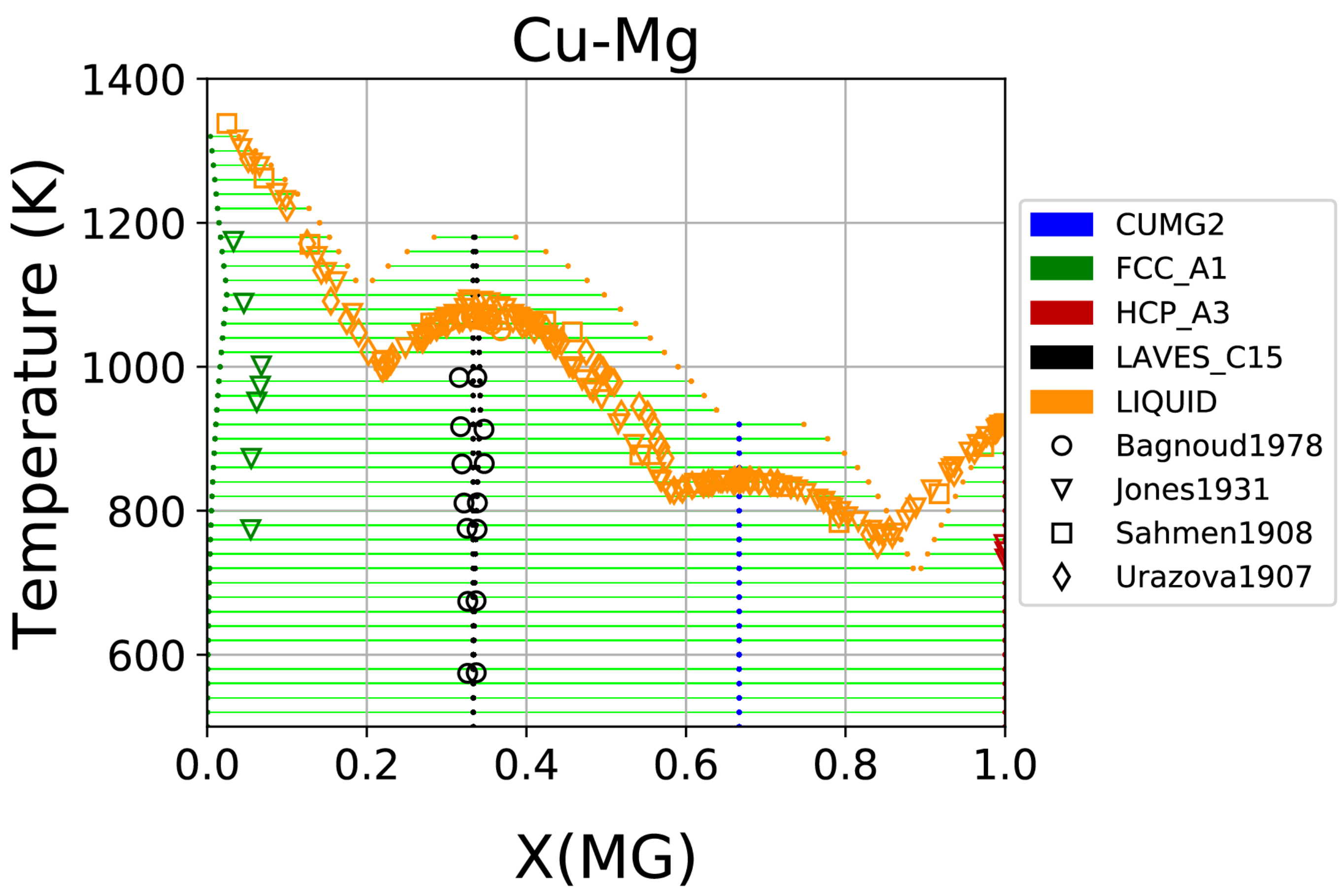
Analogous experiment: equilibrated alloy with EPMA-measured phase compositions

Parameters optimized by minimizing driving forces



Applying parameter selection to Cu-Mg

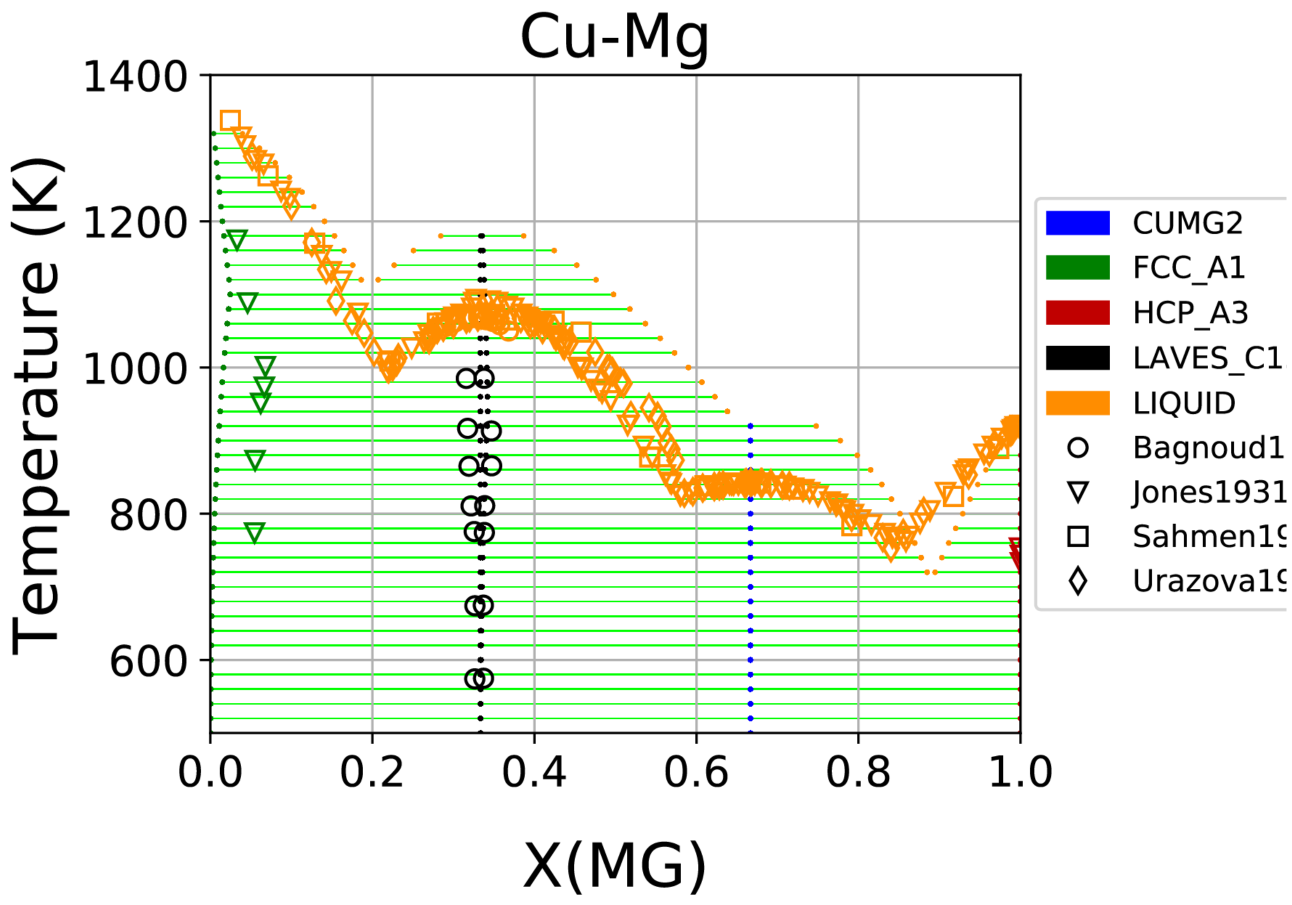
Single phase data start point



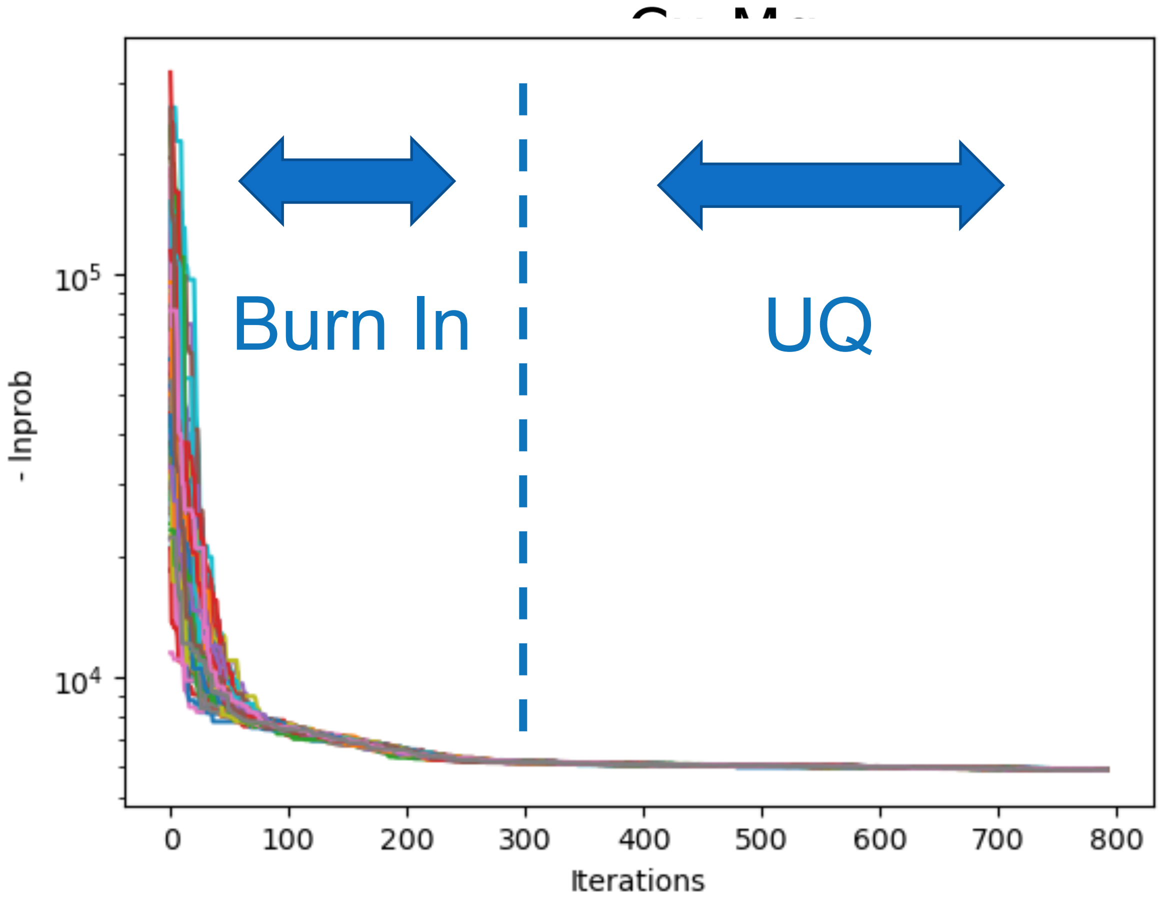
Bocklund, MRS Comm (2019)

Updating selected parameters with MCMC

Single phase data start point



MCMC to fit to thermochemical and phase equilibrium data



Bocklund, MRS Comm (2019)

Interactive Demo

Optimization and Uncertainty Quantification

ESPEI: Database Development and UQ

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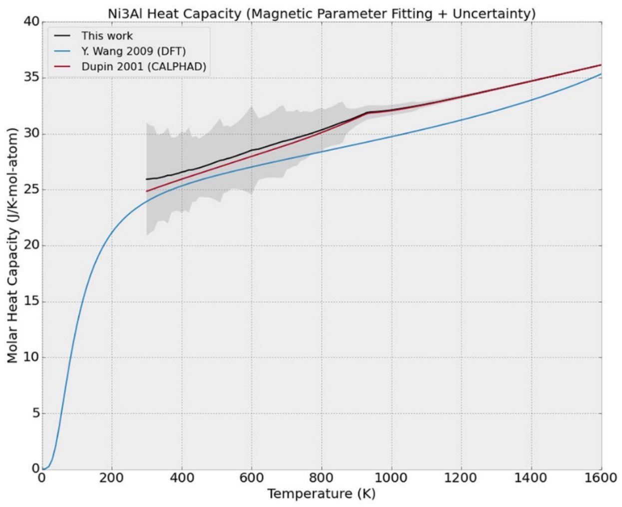
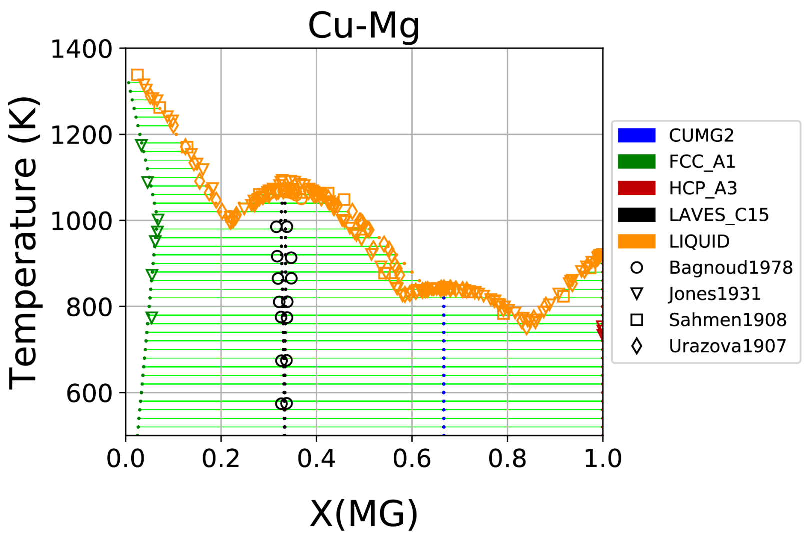
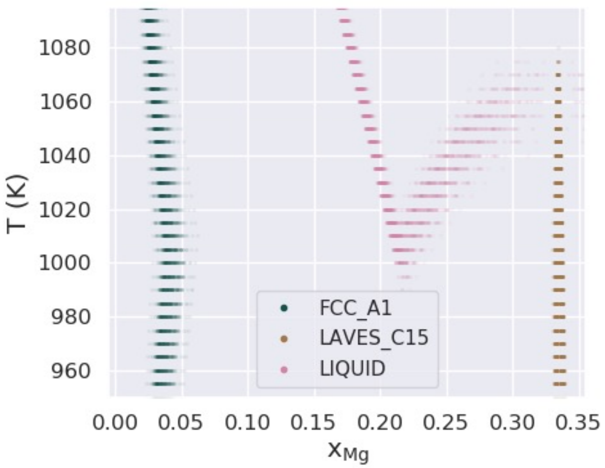
https://espei.org



Otis, Liu, JOM (2017)



Bocklund, MRS Comm (2019)



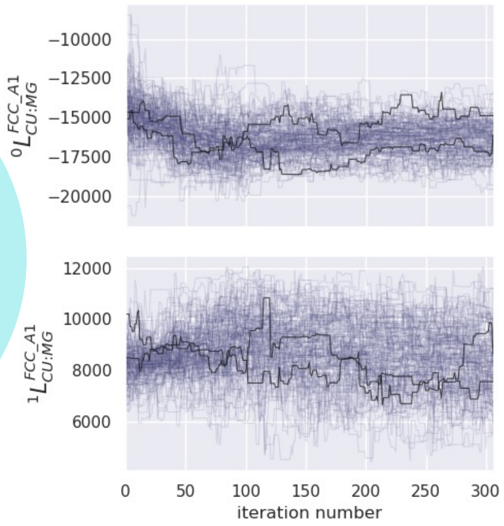
Paulson, Acta Mat. (2019)

Propagate uncertainty to thermodynamic properties

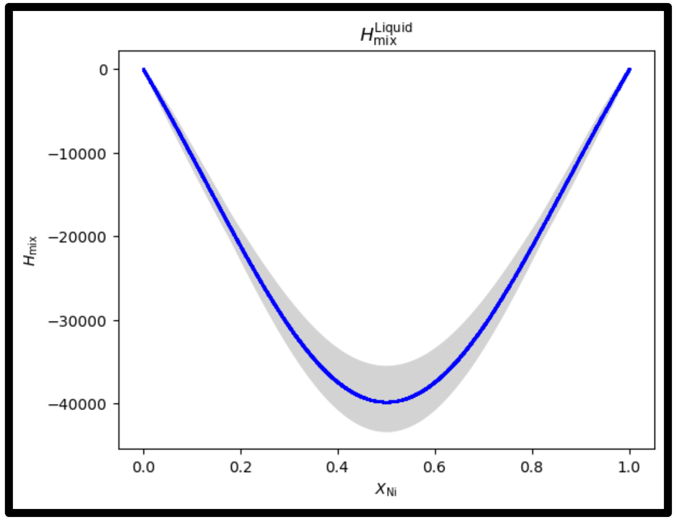
CALPHAD models

```
...
PARAMETER G(FCC_A1,CU;0)    1 GFCCCU#; 10000 N !
PARAMETER G(FCC_A1,MG;0)    1 GFCCMG#; 10000 N !
PARAMETER L(FCC_A1,CU,MG;0) 1 VV0002#; 10000 N !
PARAMETER L(FCC_A1,CU,MG;1) 1 VV0001#; 10000 N !
...
```

Parameter distribution samples from MCMC



Superimposed CALPHAD predictions: Cu-Mg

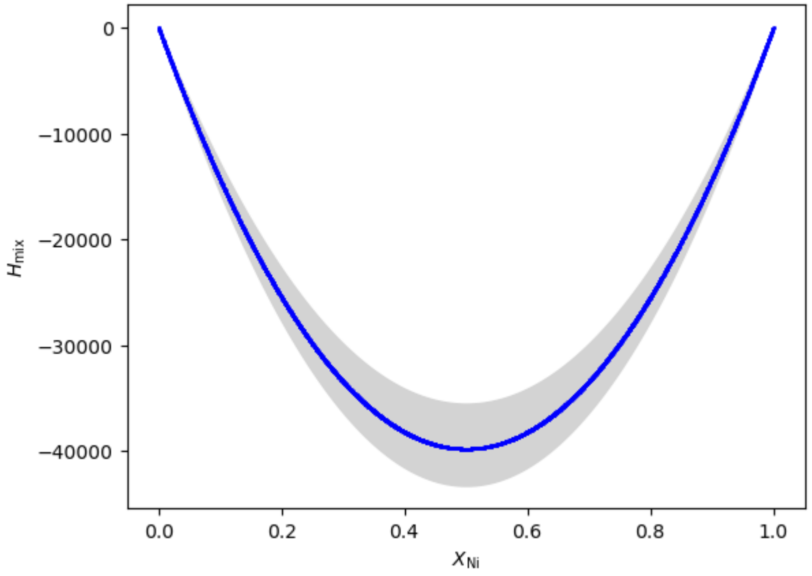


arXiv:1901.10510

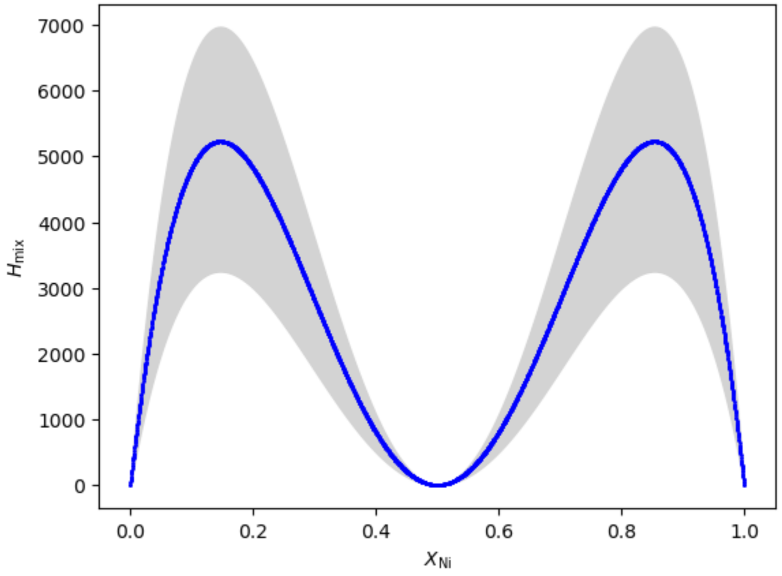
Paulson, Acta Mat. (2019)

Uncertainty quantification in liquid interaction parameters

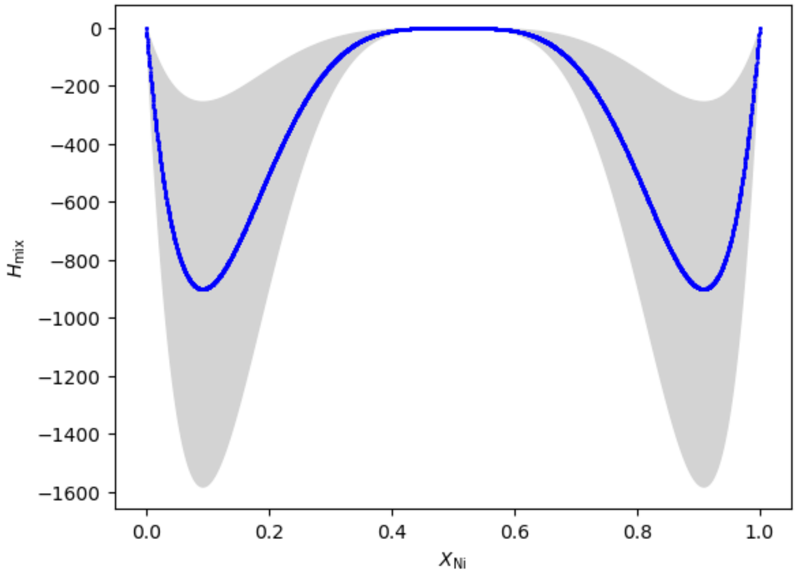
$^{0}L^{\text{liquid}}$



$^{1}L^{\text{liquid}}$



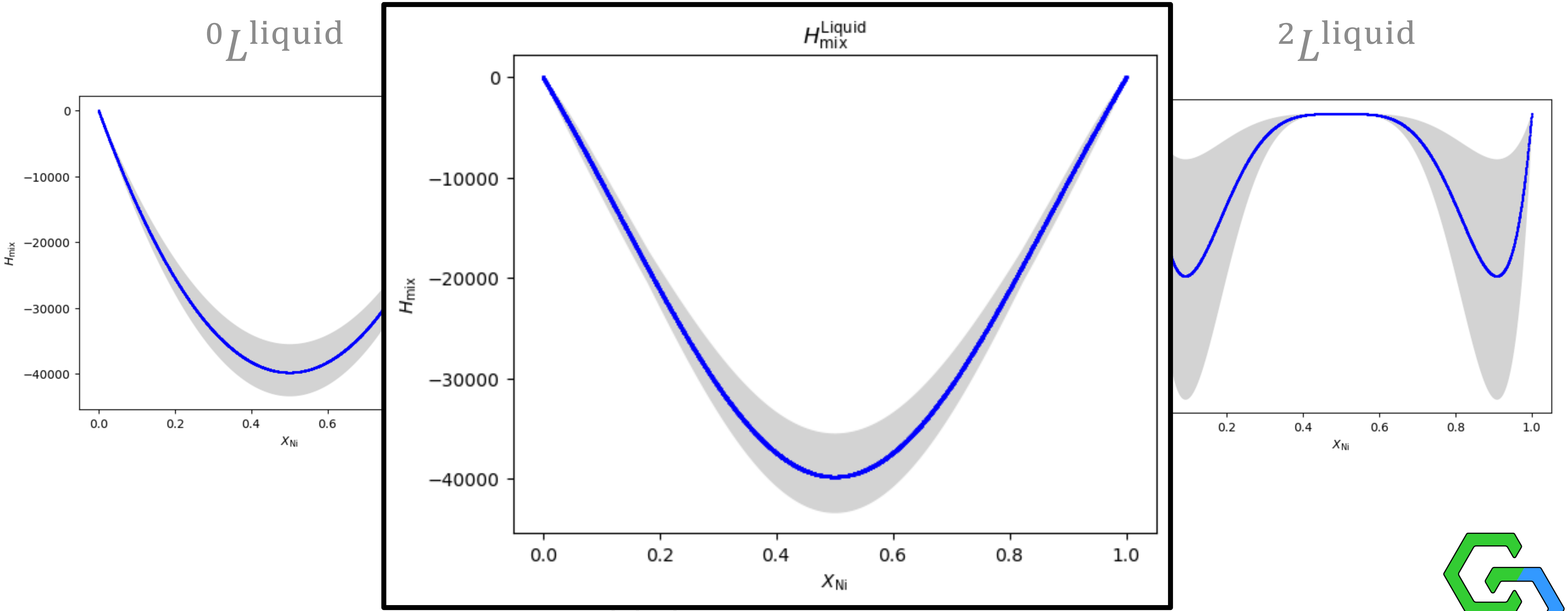
$^{2}L^{\text{liquid}}$



$$\sum_{i,j\neq i} x_i x_j \left(x_i - x_j\right)^{v}\, {}^{v}L$$

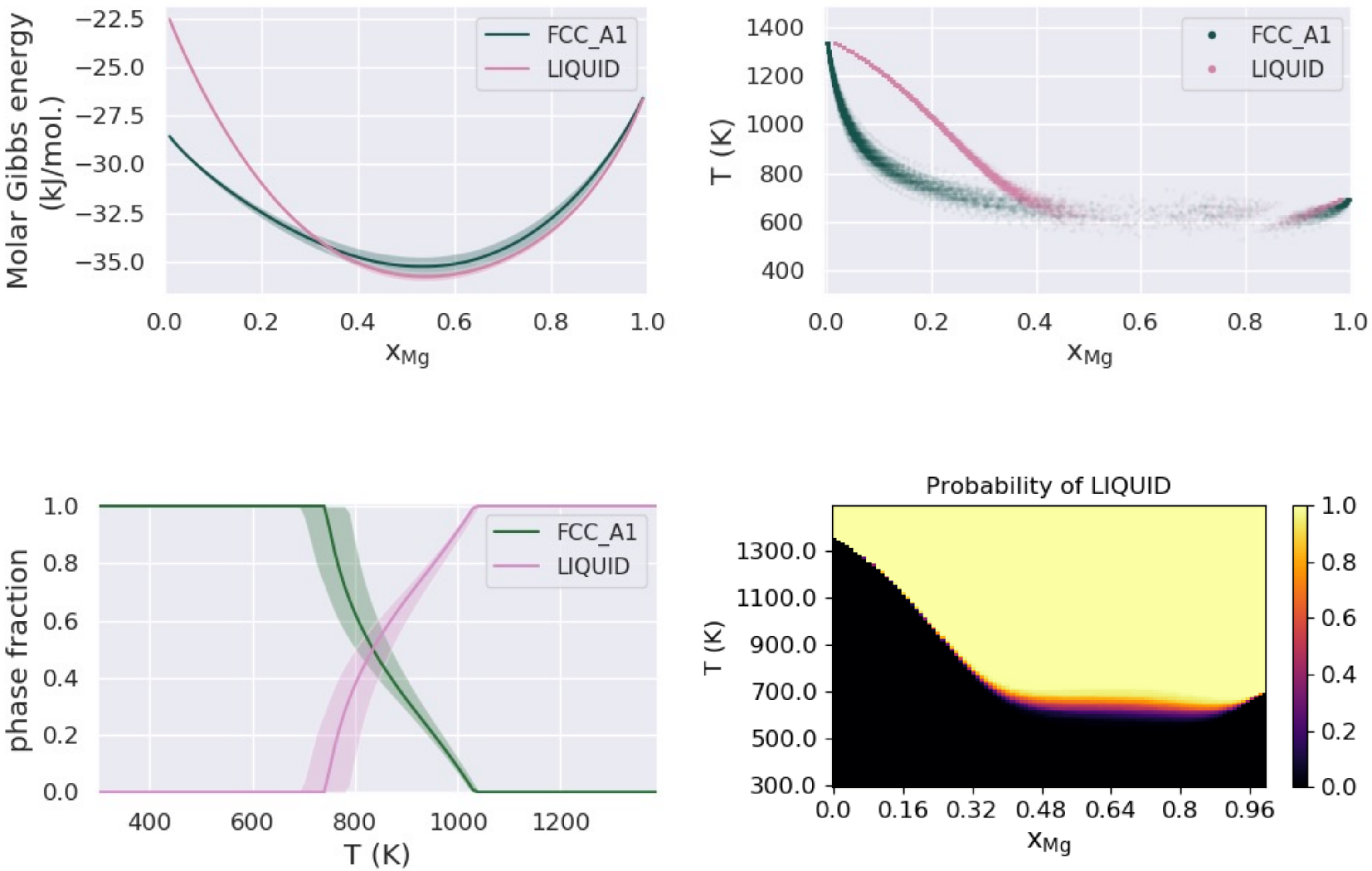
Uncertainty quantification in liquid interaction parameters

${}^{0}L^{\text{liquid}}$ ${}^{2}L^{\text{liquid}}$



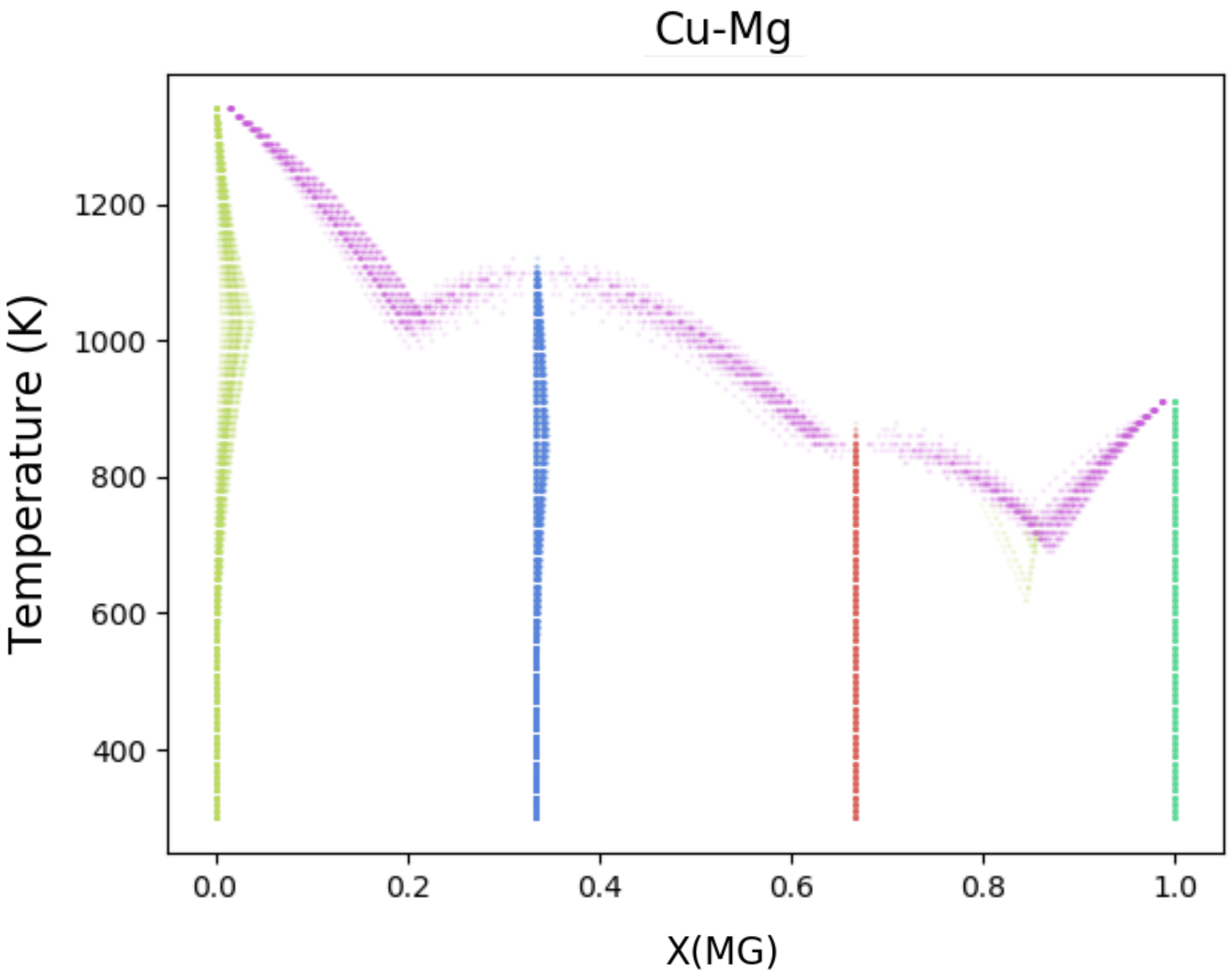
$i,j\neq i$

Propagation of uncertainty to calculated properties and phase diagrams



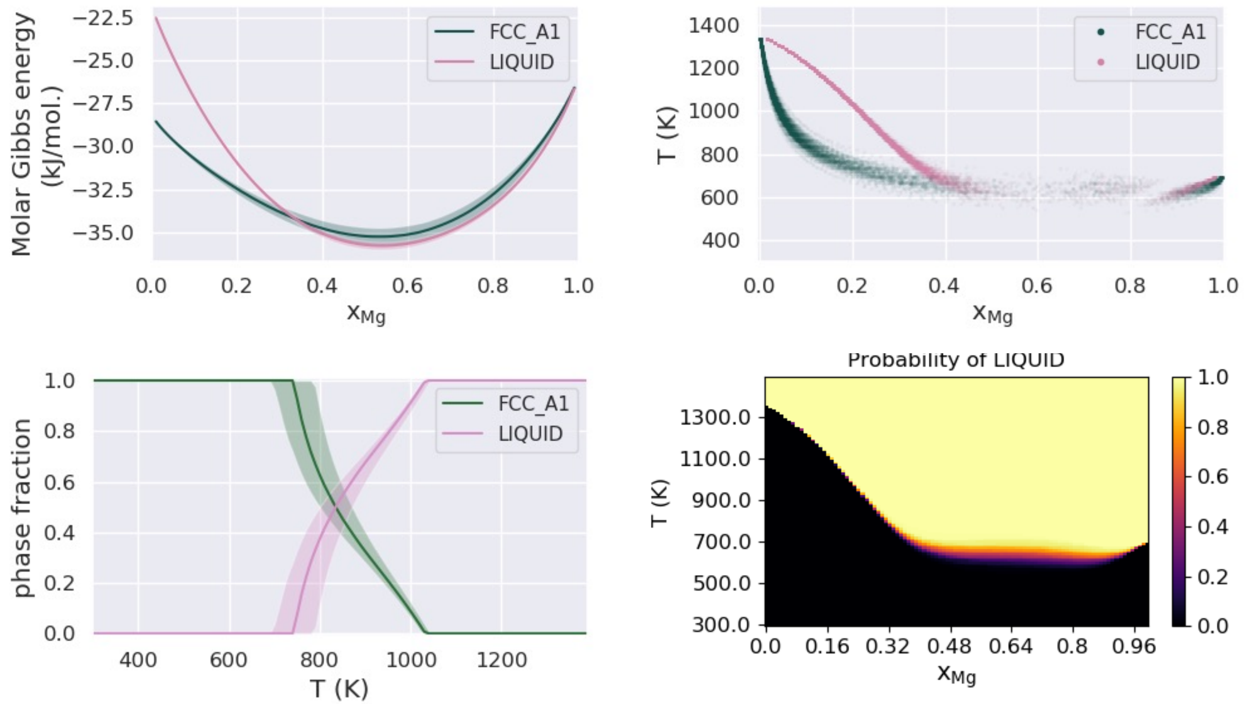
Paulson, Acta Mat. (2019)

Propagation of uncertainty to calculated properties and phase diagrams



Paulson, Acta Mat. (2019)

PDUQ: Phase Diagram Uncertainty Quantification package



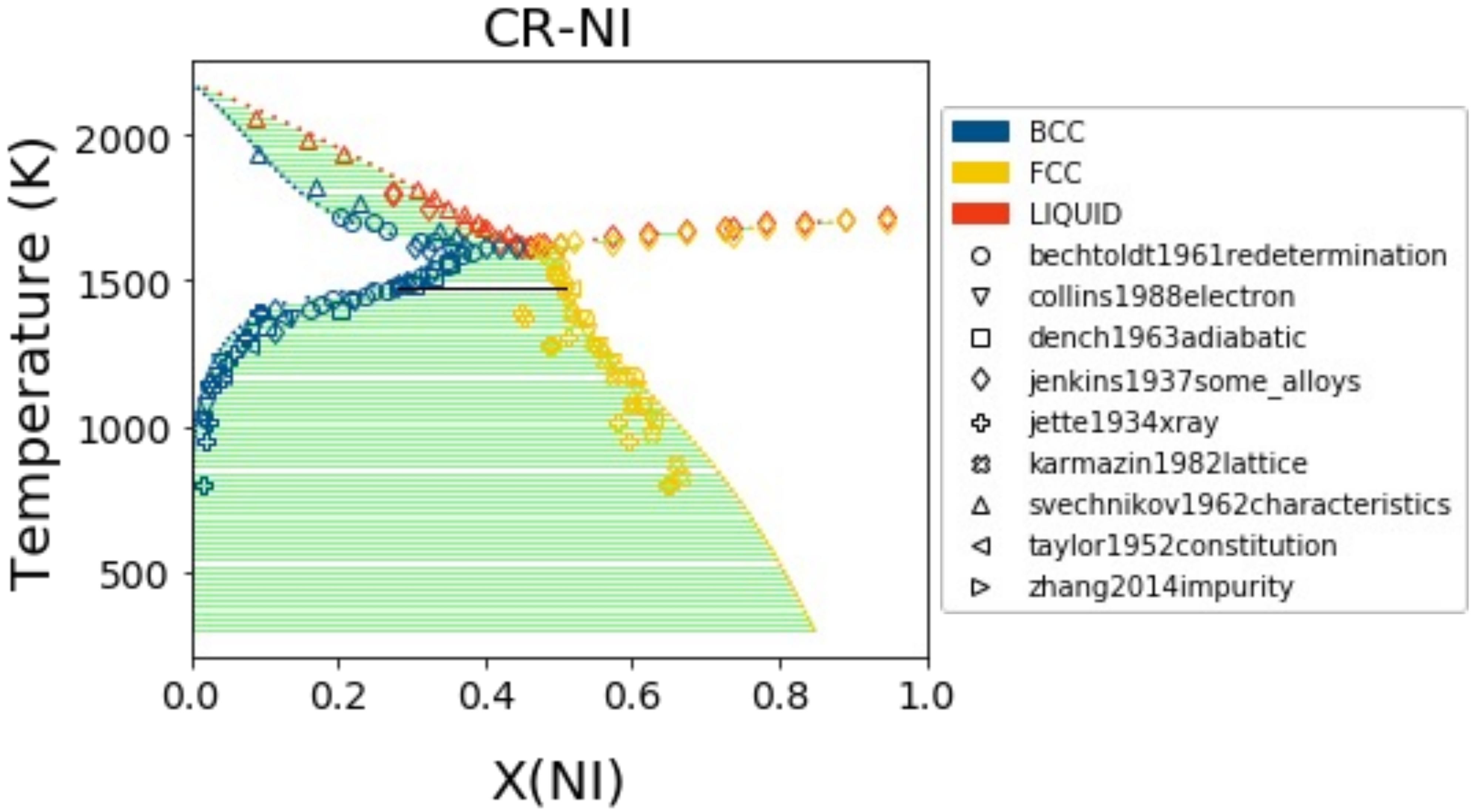
Noah Paulson

Marius Stan

Argonne National Lab

https://pduq.readthedocs.io

Cr-Ni Phase Diagram (2000 MCMC iterations)



Journal of MATERIALS RESEARCH

Invited Feature Paper
DOI: 10.1557/jmr.2020.269

https://doi.org/10.1557/jmr.2020.269

Sensitivity estimation for calculated phase equilibria

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[2]*Department of Materials Science and Engineering, Pennsylvania State University, University Park, Pennsylvania 16802, USA*
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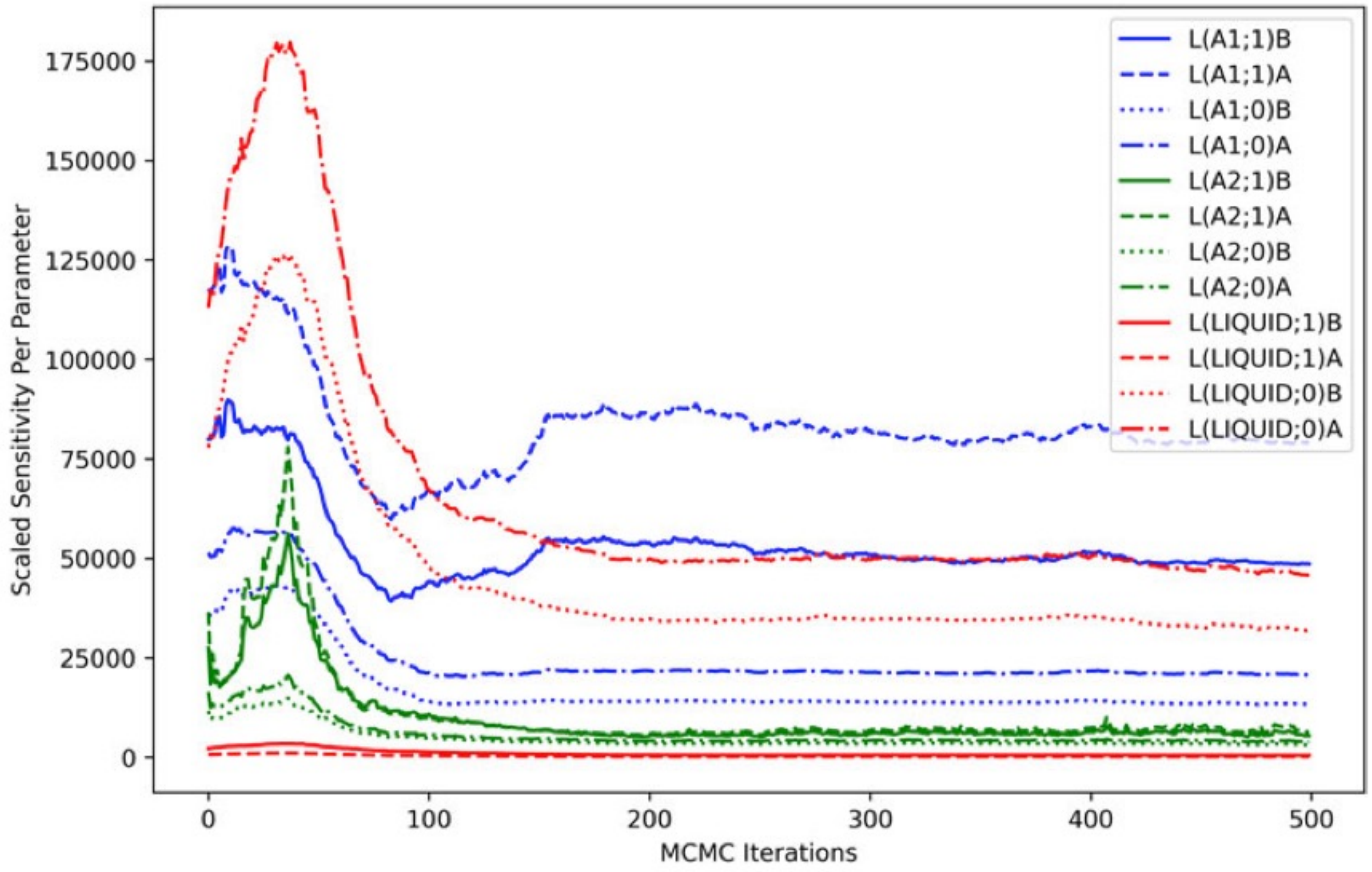
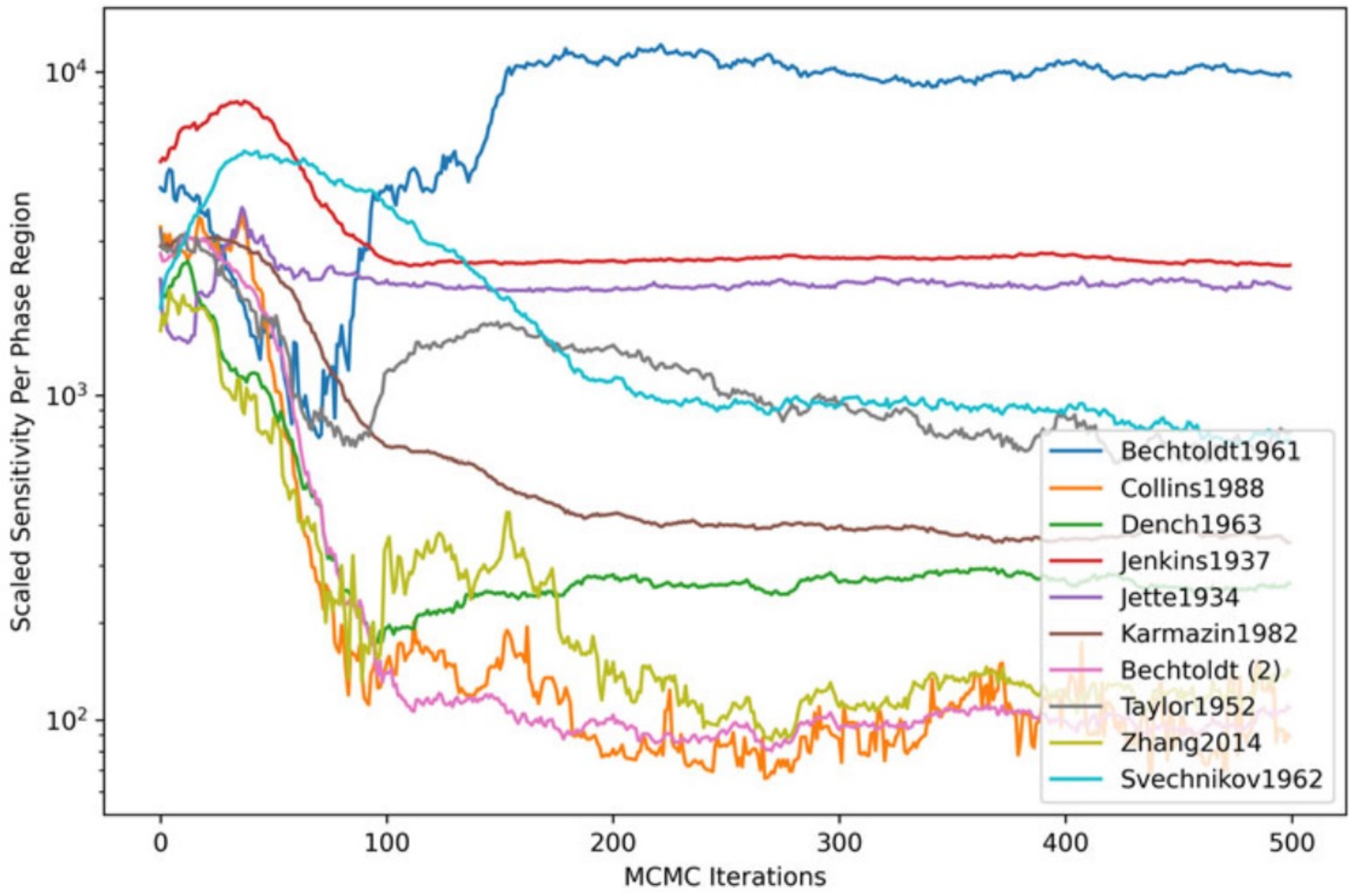
The development of a consistent framework for Calphad model sensitivity is necessary for the rational reduction of uncertainty via new models and experiments. In the present work, a sensitivity theory for Calphad was developed, and a closed-form expression for the log-likelihood gradient and Hessian of a multi-phase equilibrium measurement was presented. The inherent locality of the defined sensitivity metric was mitigated through the use of Monte Carlo averaging. A case study of the Cr–Ni system was used to demonstrate visualizations and analyses enabled by the developed theory. Criteria based on the classical Cramér–Rao bound were shown to be a useful diagnostic in assessing the accuracy of parameter covariance estimates from Markov Chain Monte Carlo. The developed sensitivity framework was applied to estimate the statistical value of phase equilibria measurements in comparison with thermochemical measurements, with implications for Calphad model uncertainty reduction.

Cr-Ni Parameter Sensitivity



Otis, JMR (2020)

Cr-Ni Dataset Sensitivity



Otis, JMR (2020)

Interactive Demo

Uncertainty Propagation

Summary of Notable Features

- Parameter generation
  - Generate multicomponent CEF models with Redlich-Kister polynomials from thermochemical data
  - Select optimal model forms based on statistical information criteria
- MCMC optimization, UQ, and UP
  - Optimize and quantify uncertainty for any PyCalphad model
  - Software infrastructure designed to be extendable to new types of data and different optimization and UQ backends
  - HPC capable
- Command line interface and interactive Python API are supported

Getting help

- PyCalphad
  - https://pycalphad.org
  - Gitter channel (chat)
    - https://gitter.im/pycalphad/pycalphad
  - GitHub Discussions (forums)
    - https://github.com/pycalphad/pycalphad/discussions
- ESPEI
  - https://espei.org
  - Gitter channel (chat)
    - https://gitter.im/PhasesResearchLab/ESPEI
  - GitHub Discussions (forums)
    - https://github.com/phasesresearchlab/espei/discussions

Materials
Genome
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