Day 2: ESPEI

Database development and uncertainty quantification

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June 28-29, 2021

PyCalphad/ESPEI Virtual Workshop

CALPHAD modeling

Derivatives of Gibbs energy
enthalpy, entropy, heat capacity

Phase equilibrium data
phase boundaries

↓

Gibbs Energy of Individual Phases

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

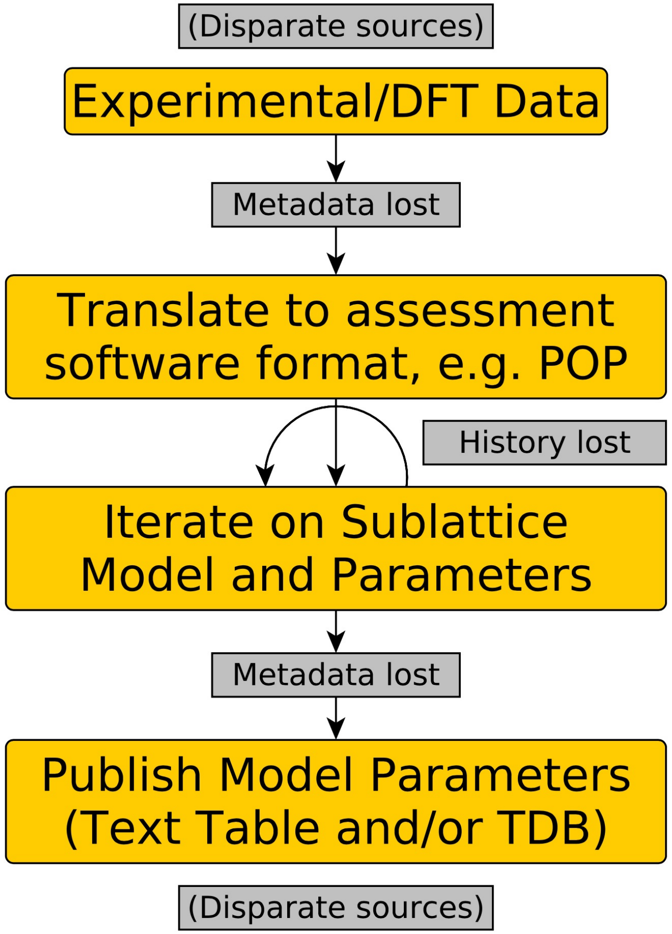
↓

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

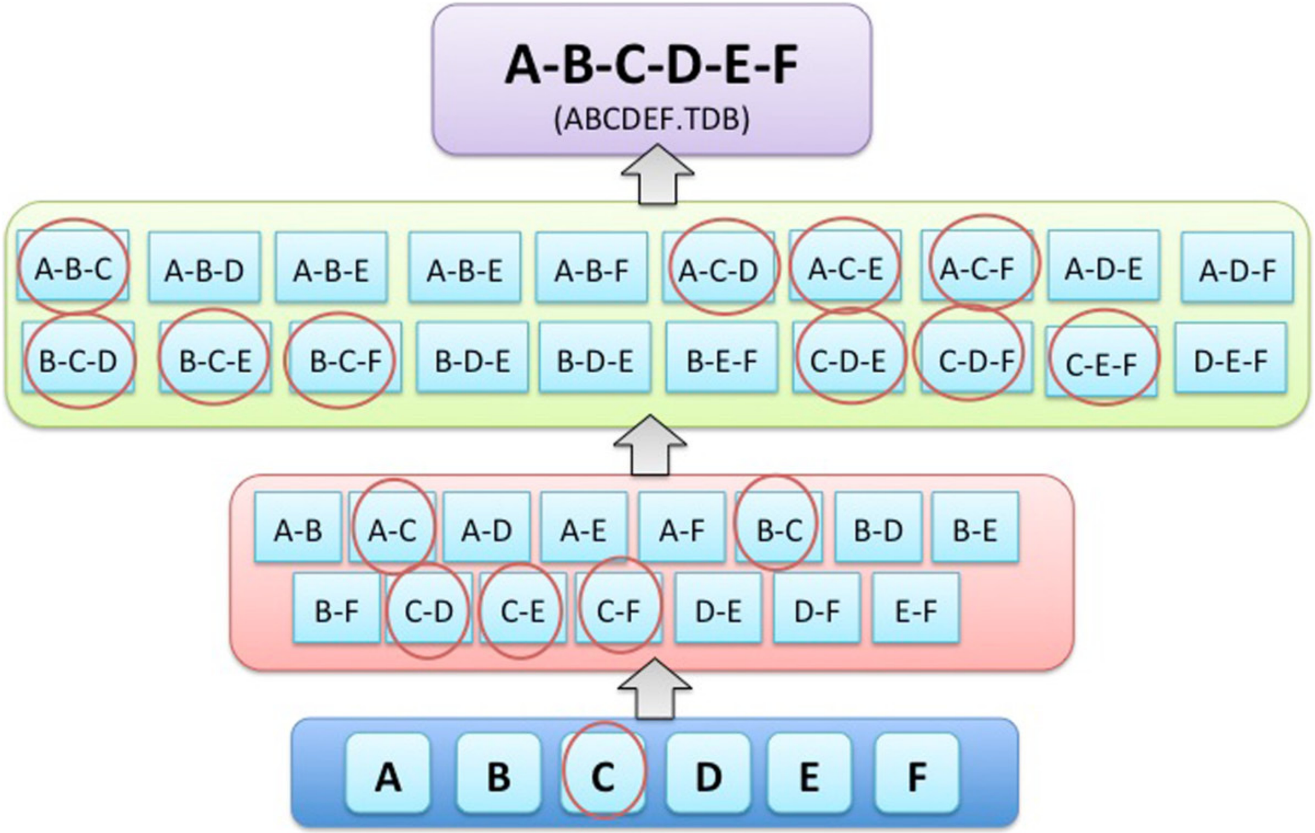
Pure elements → Binary → Ternary → Multicomponent

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

CALPHAD database development process



Otis, PhD Thesis (2016)

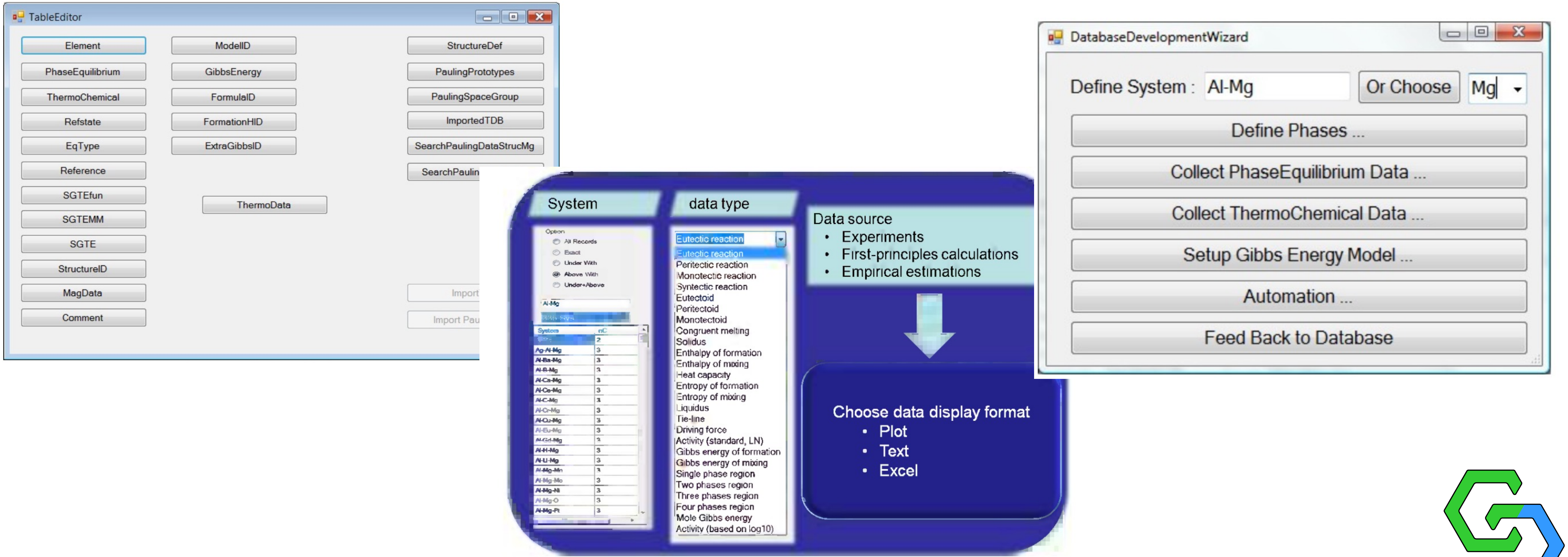


Campbell, IMMI (2014)

Magnesium Technology 2010
Edited by: Sean R. Agnew, Neale R. Neelameggham, Eric A. Nyberg, and Wim H. Sillekens
TMS (The Minerals, Metals & Materials Society), 2010

ESPEI: Extensible, Self-optimizing Phase Equilibrium Infrastructure for Magnesium Alloys

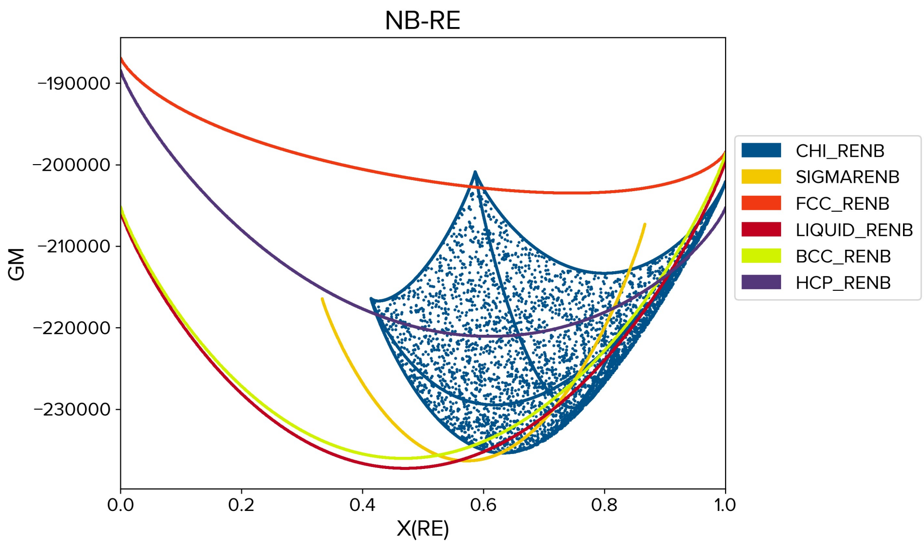
Shun-Li Shang, Yi Wang, and Zi-Kui Liu
MaterialsInformatics LLC, State College, Pennsylvania 16803, USA

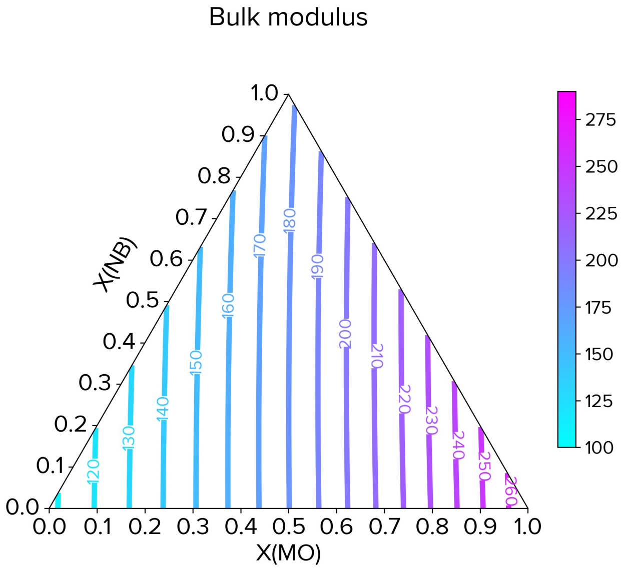


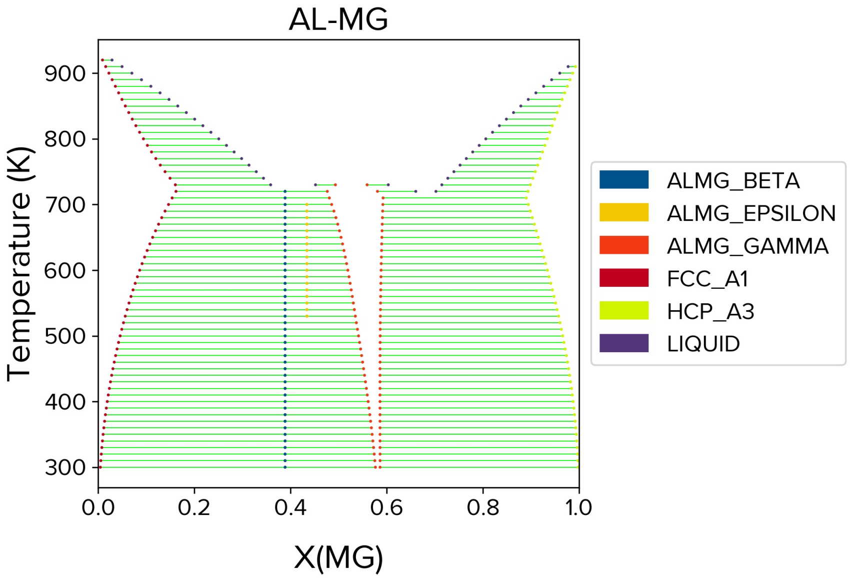
pycalphad: open-source Calphad library

- 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







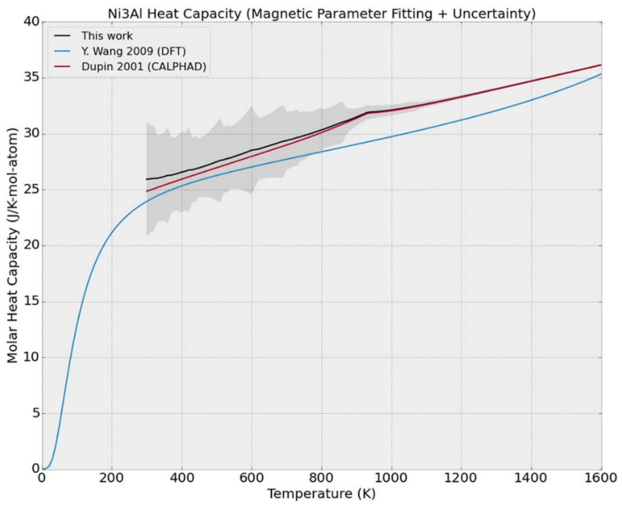
https://pycalphad.org

ESPEI: Database Development and UQ

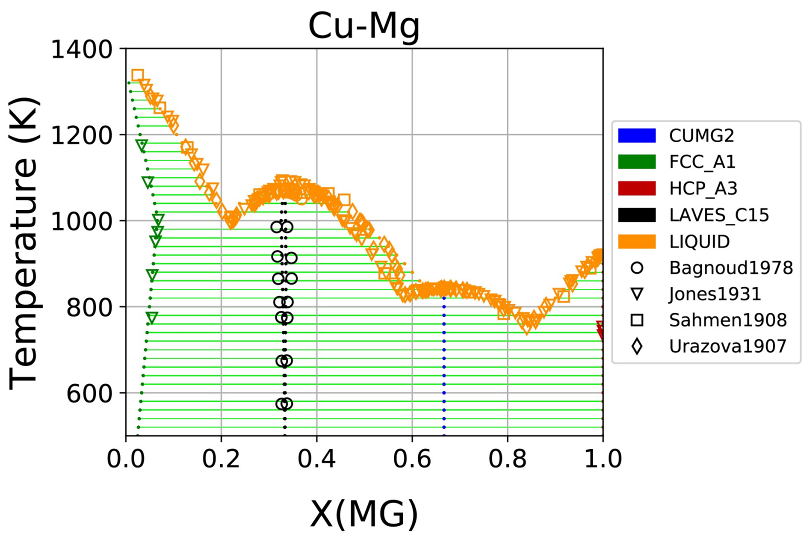
Extensible Self-optimizing Phase Equilibria Infrastructure

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

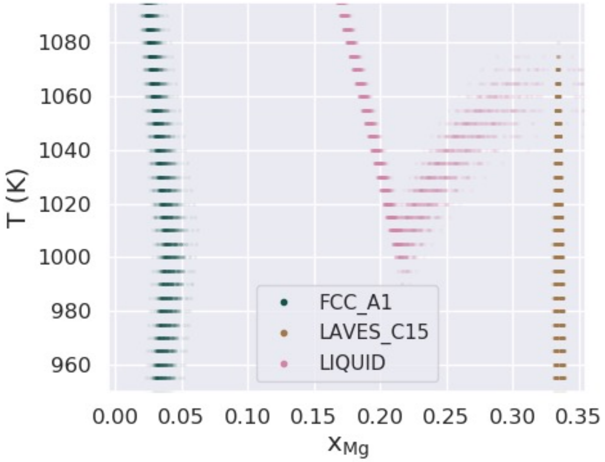
https://espei.org



Otis, Liu, JOM (2017)

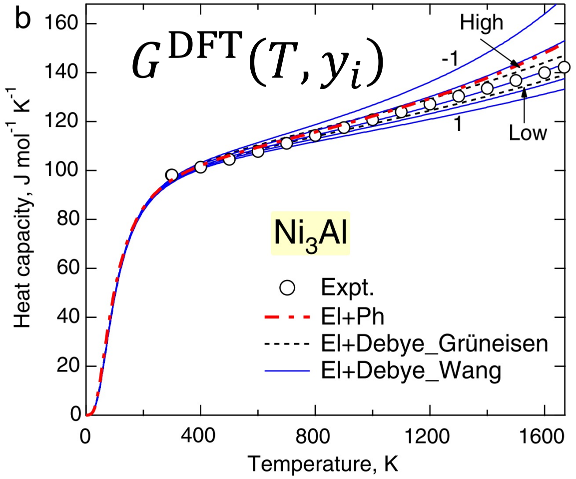


Bocklund, MRS Comm (2019)



Paulson, Acta Mat. (2019)

Parameter selection in ESPEI



Shang, Comput. Mater. Sci. (2009)

Fit non-equilibrium thermochemical data to a functional form

Temperature dependence: power expansion

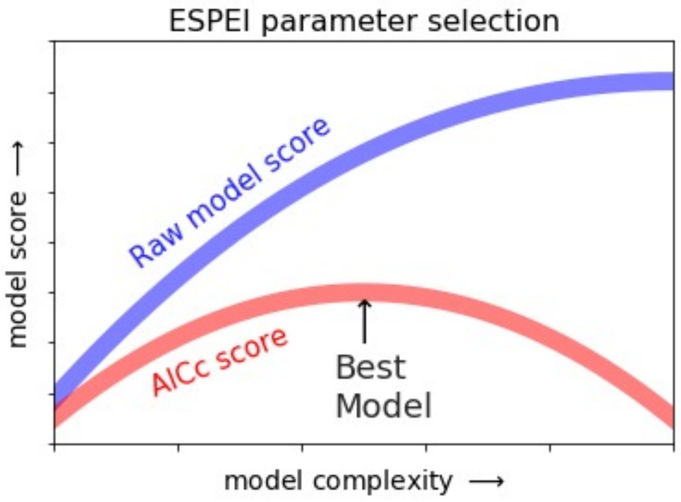
$$G(T) = a + b\,T + c\,T\,\ln T + d\,T^2 + e\,T^{-1}$$

Composition dependence: Redlich-Kister polynomial

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

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

Overfitting prevented with corrected Akaike information criterion

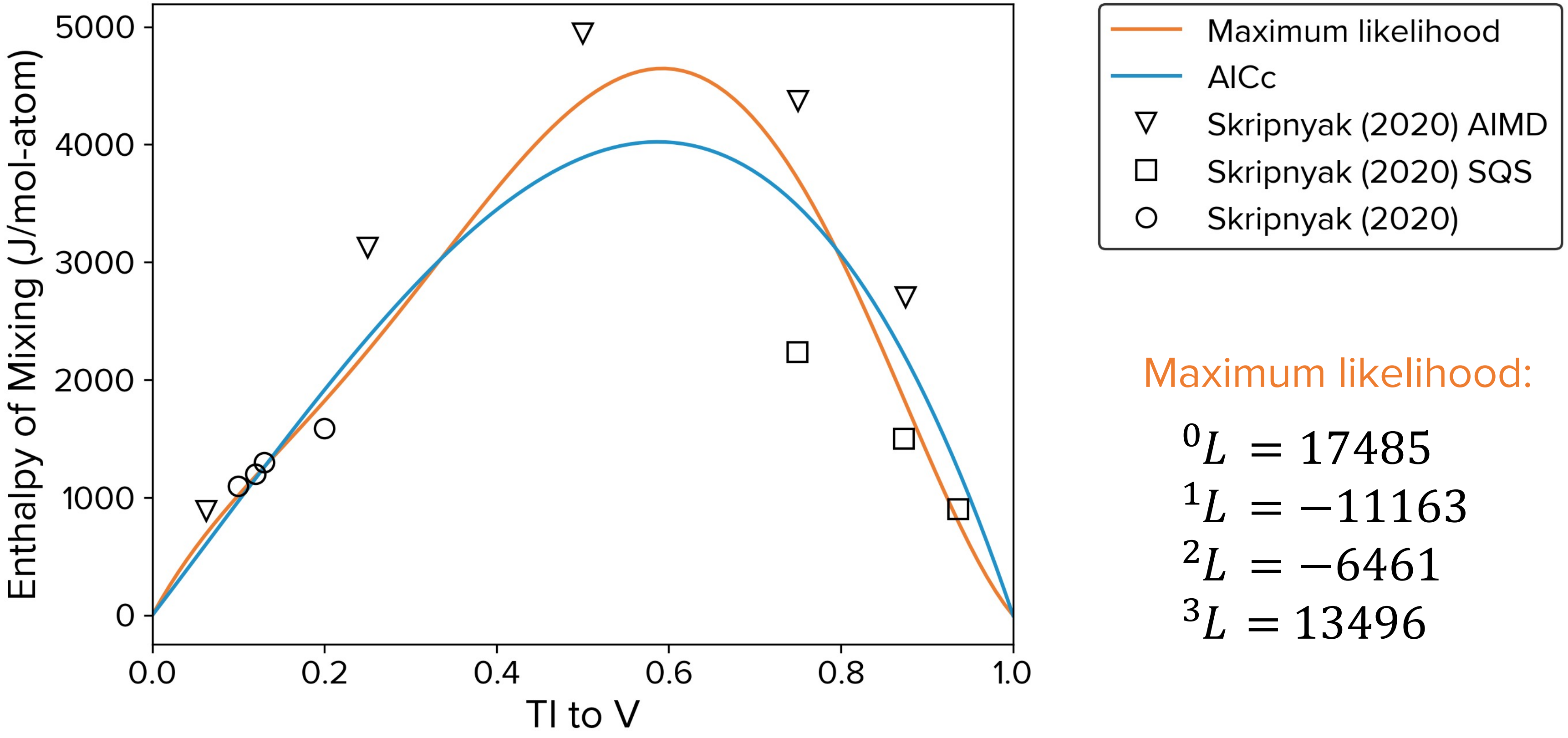


VanderPlas (2016)

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

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

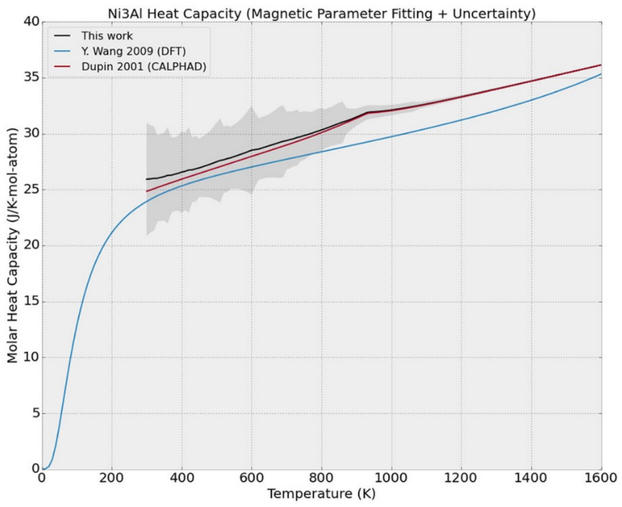
ESPEI Parameter Generation

ESPEI: Database Development and UQ

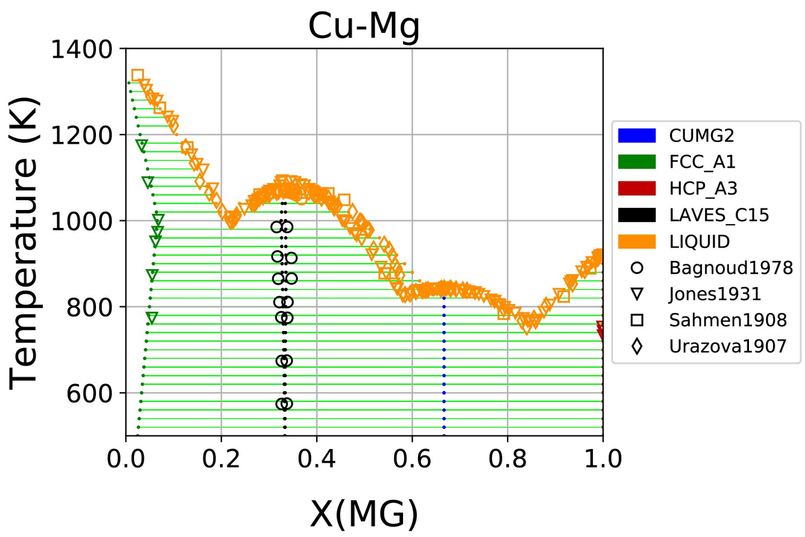
Extensible Self-optimizing Phase Equilibria Infrastructure

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

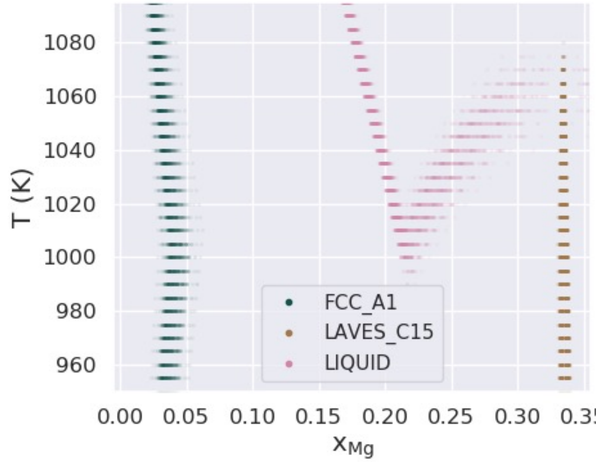
https://espei.org



Otis, Liu, JOM (2017)



Bocklund, MRS Comm (2019)



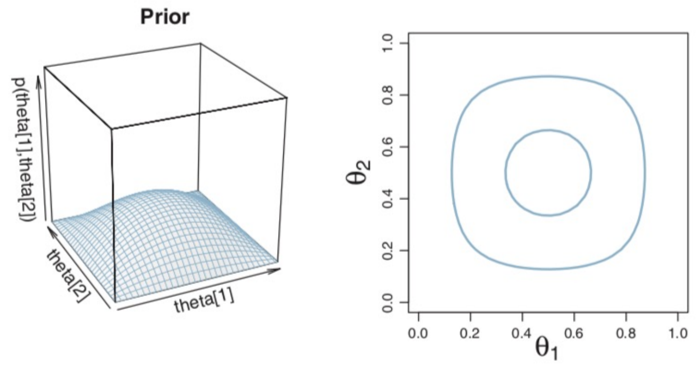
Paulson, Acta Mat. (2019)

MCMC: Bayesian parameter estimation

Posterior	Likelihood	Prior

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

Data

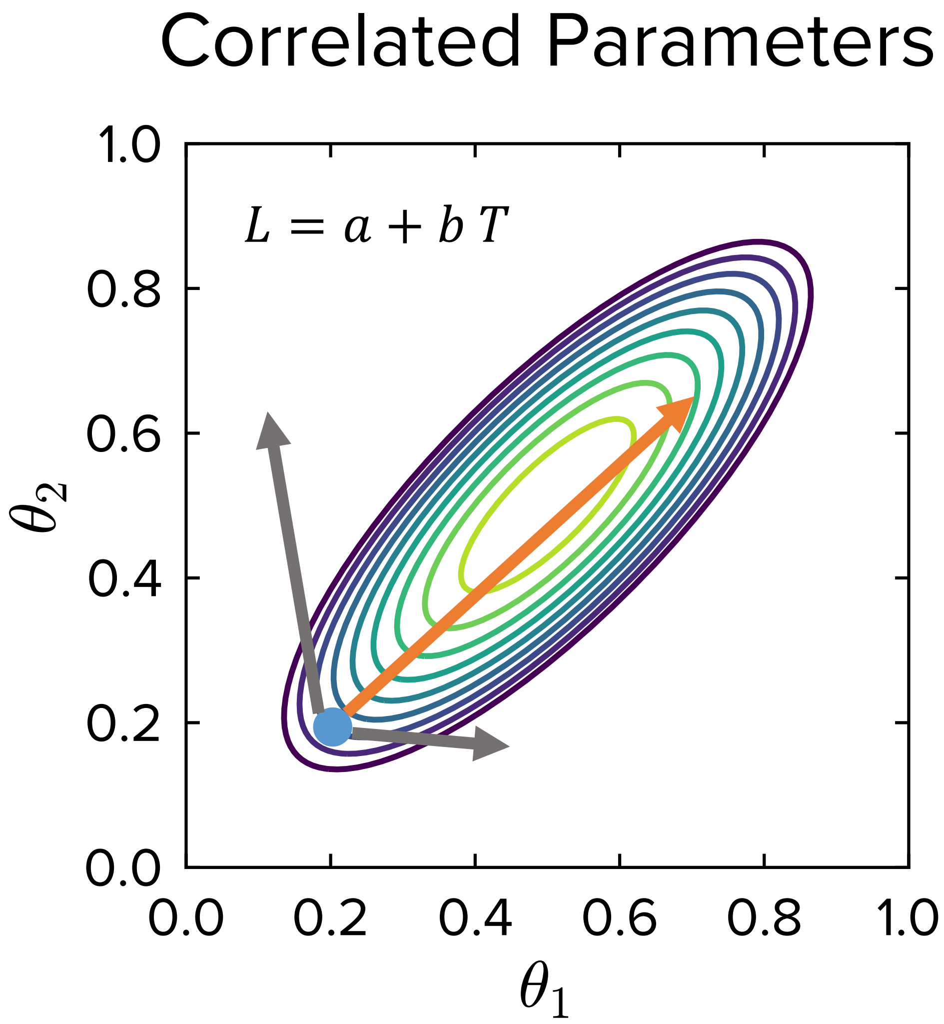


- Markov chain
  - Sequence of values that are independent from each other
- Monte Carlo
  - Random sampling

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)

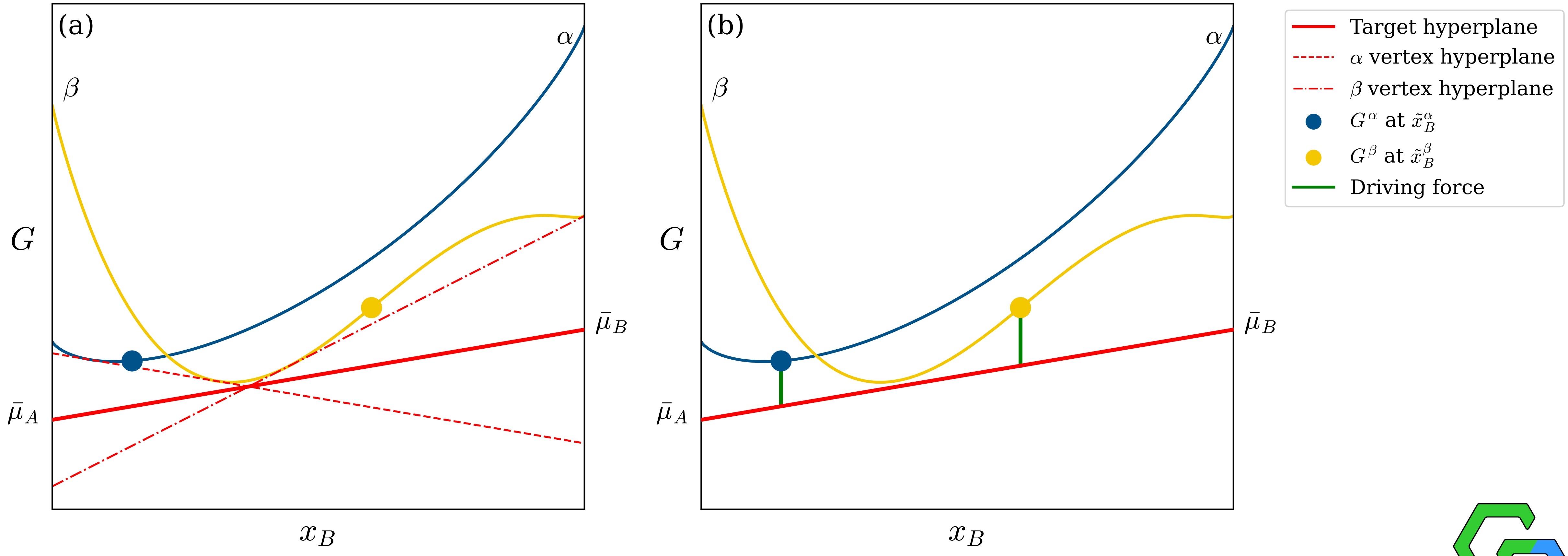
Ensemble MCMC: efficient exploration of correlated parameter space



- 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

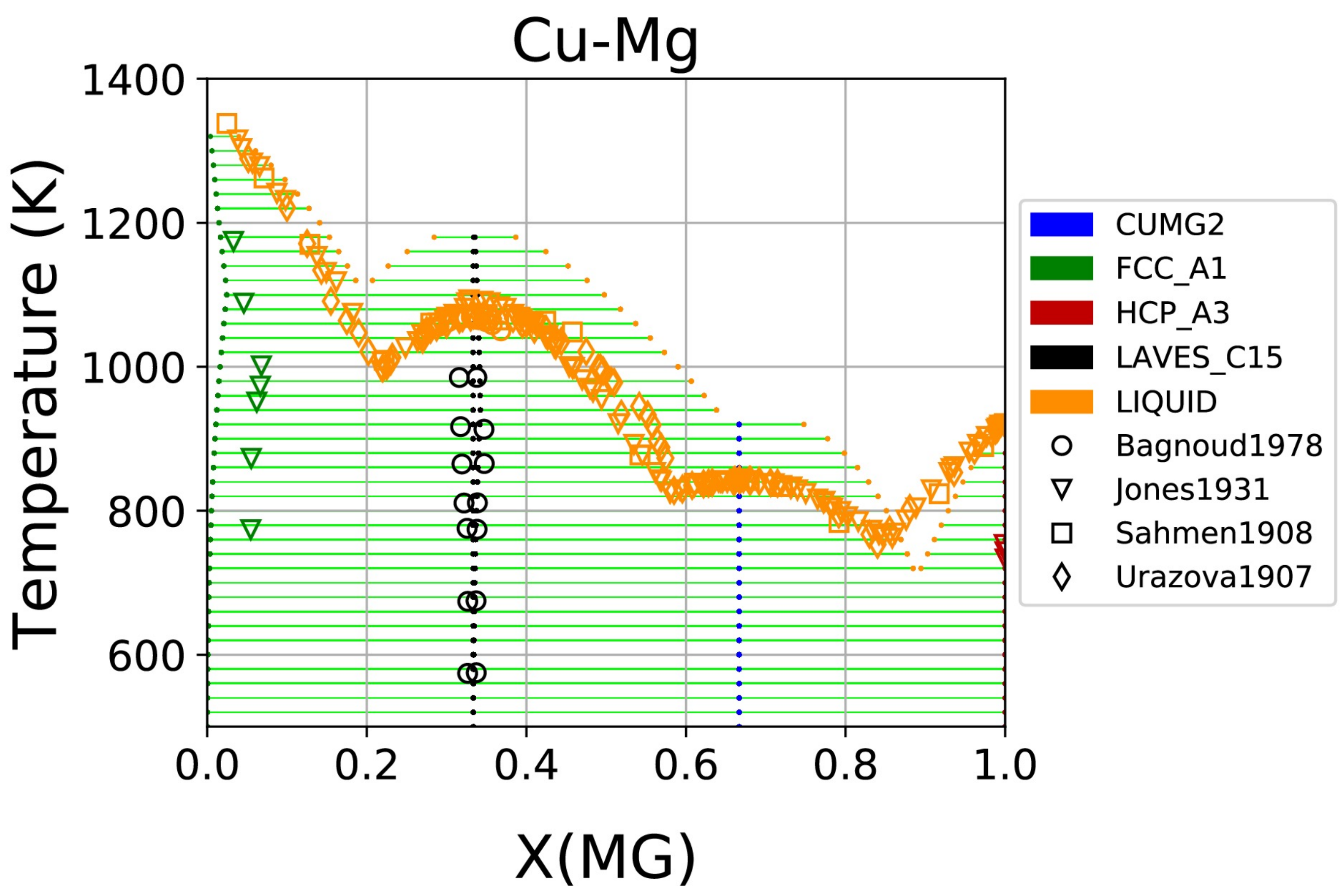
Goal: Mean hyperplane matches experimental phase compositions



Bocklund, MRS Comm (2019)

Applying parameter selection to Cu-Mg

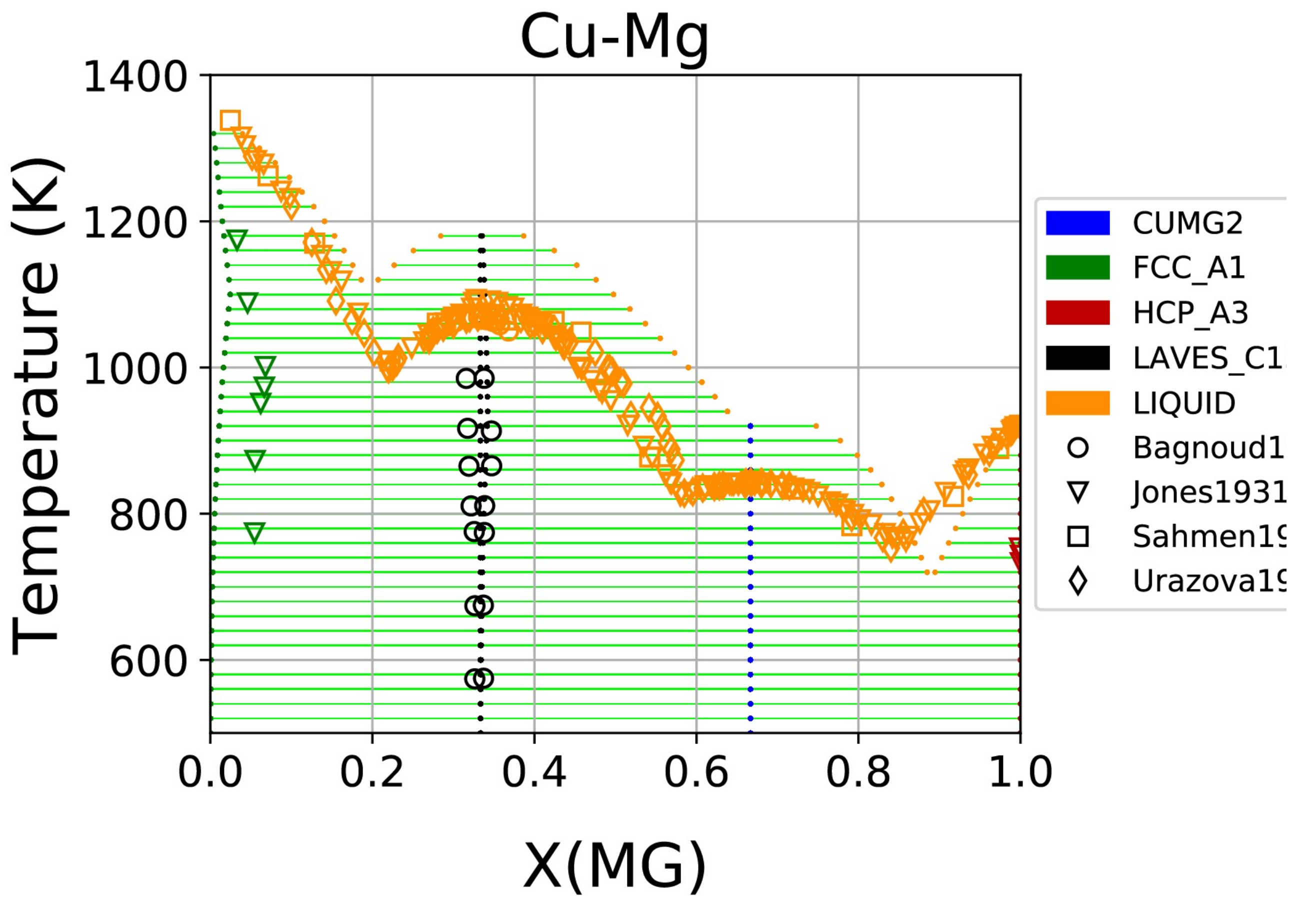
Single phase data start point



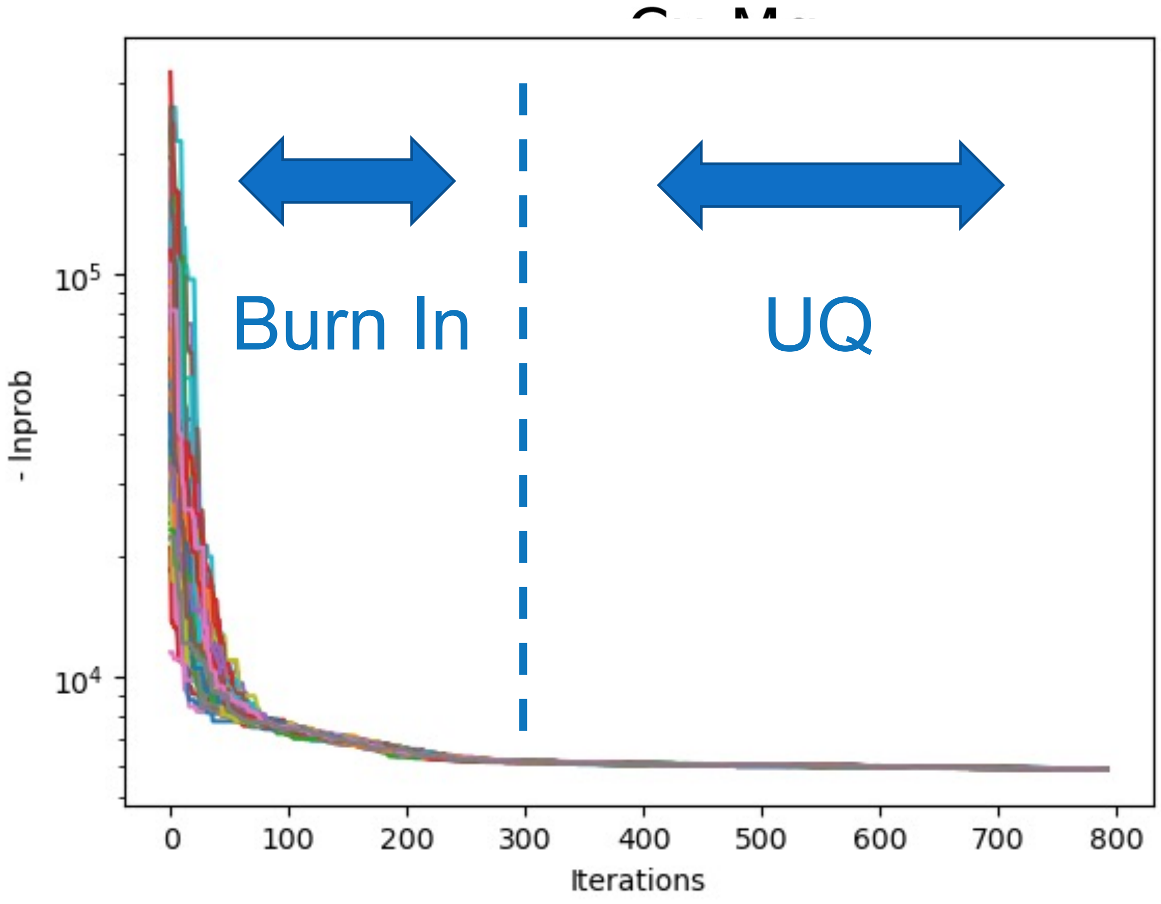
Bocklund, MRS Comm (2019)

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

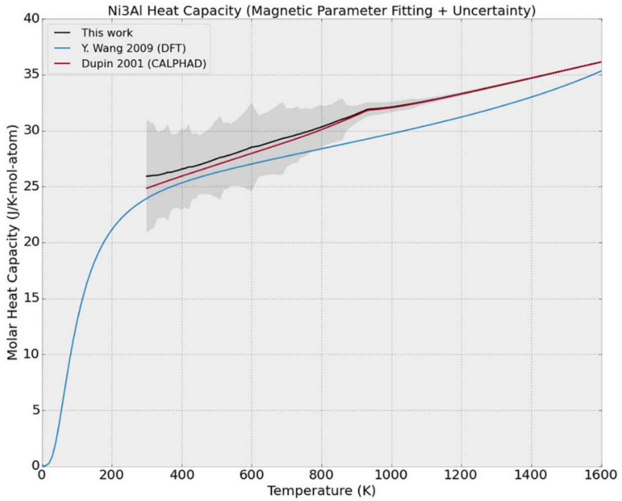
MCMC Optimization and Uncertainty Quantification

ESPEI: Database Development and UQ

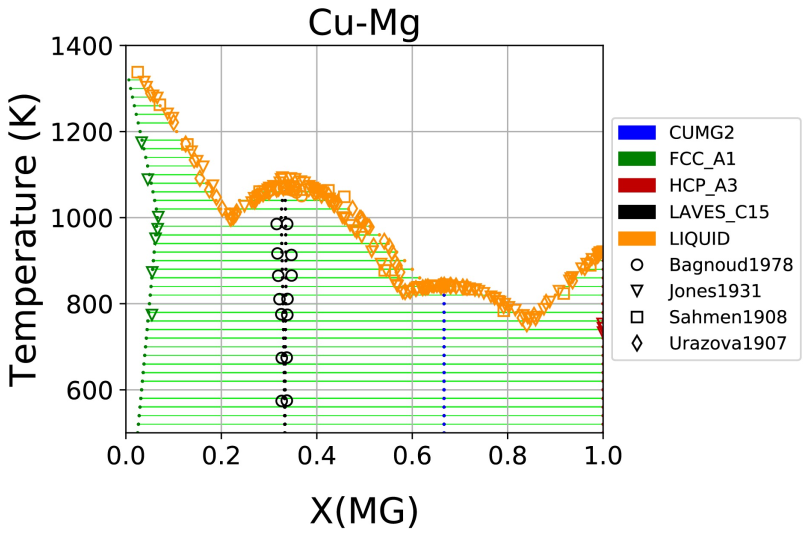
Extensible Self-optimizing Phase Equilibria Infrastructure

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

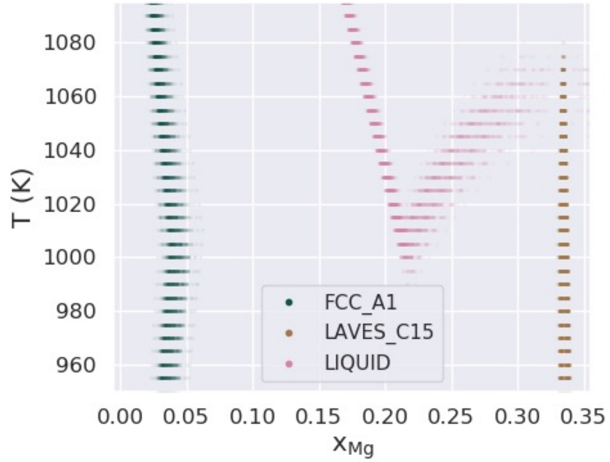
https://espei.org



Otis, Liu, JOM (2017)



Bocklund, MRS Comm (2019)



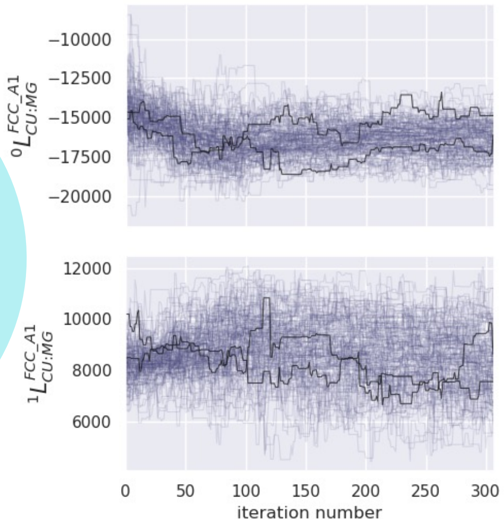
Paulson, Acta Mat. (2019)

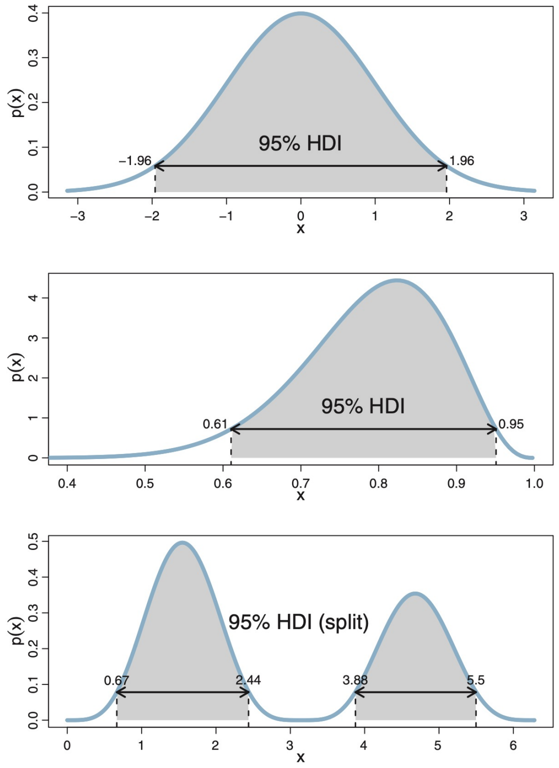
Propagate uncertainty to thermodynamic properties

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 !
...
```

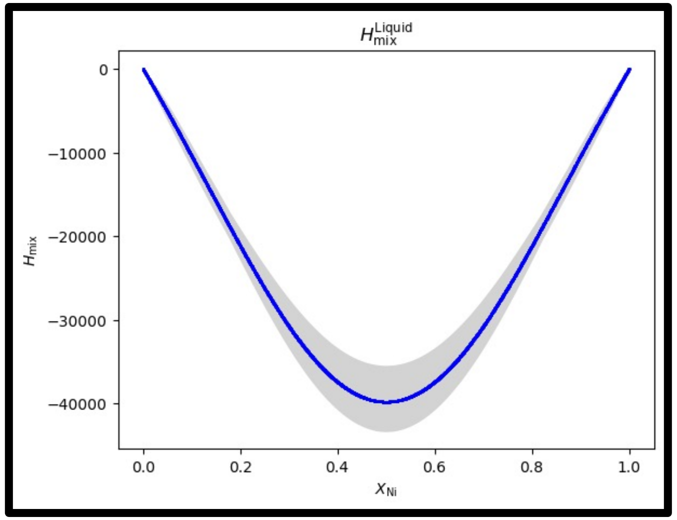
Parameter distribution samples from MCMC





Kruschke (2015)

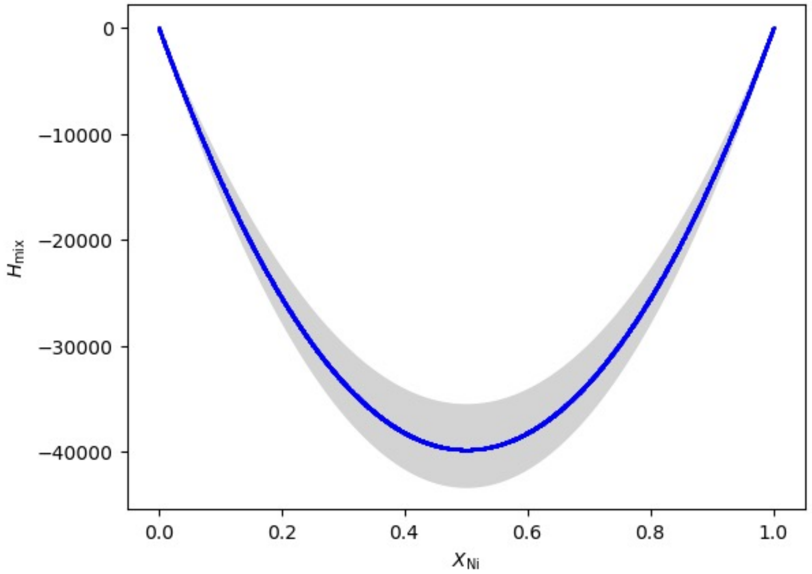
Superimposed CALPHAD predictions: Cu-Mg



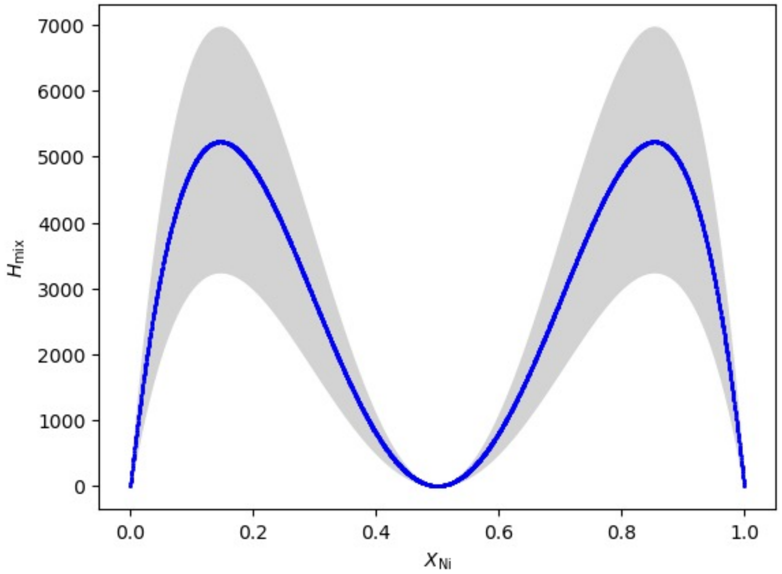
Paulson, Acta Mat. (2019)

Uncertainty quantification in liquid interaction parameters

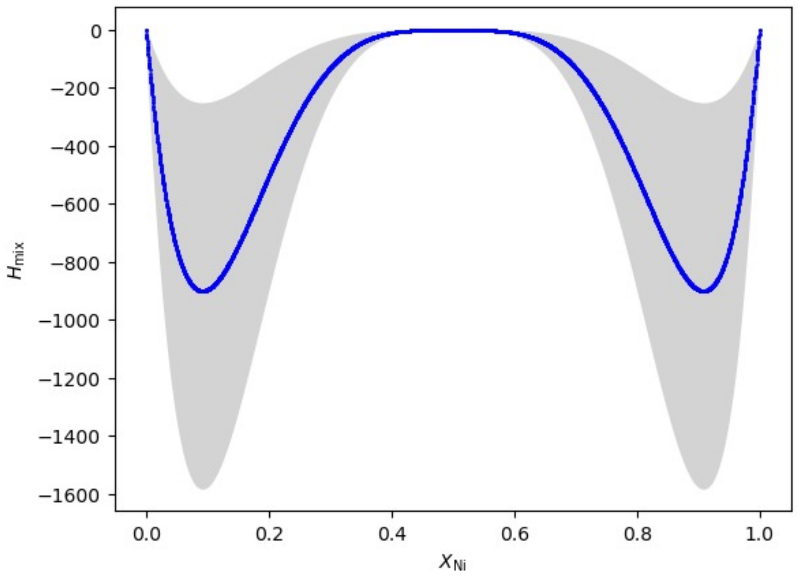
${}^{0}L^{\mathrm{liquid}}$



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



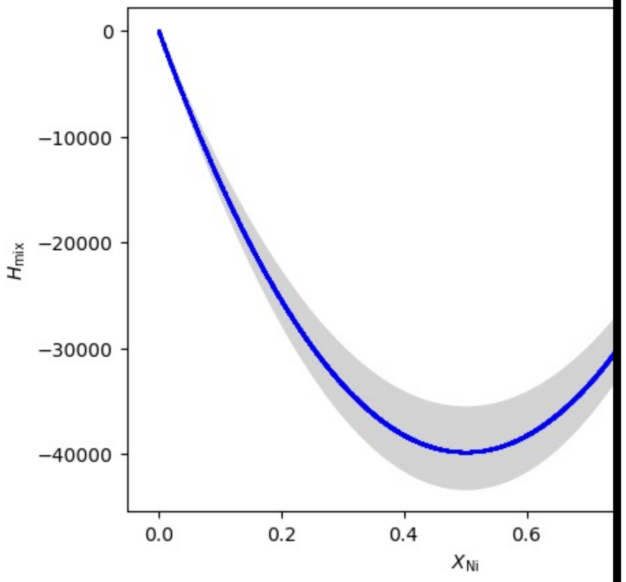
${}^{2}L^{\mathrm{liquid}}$

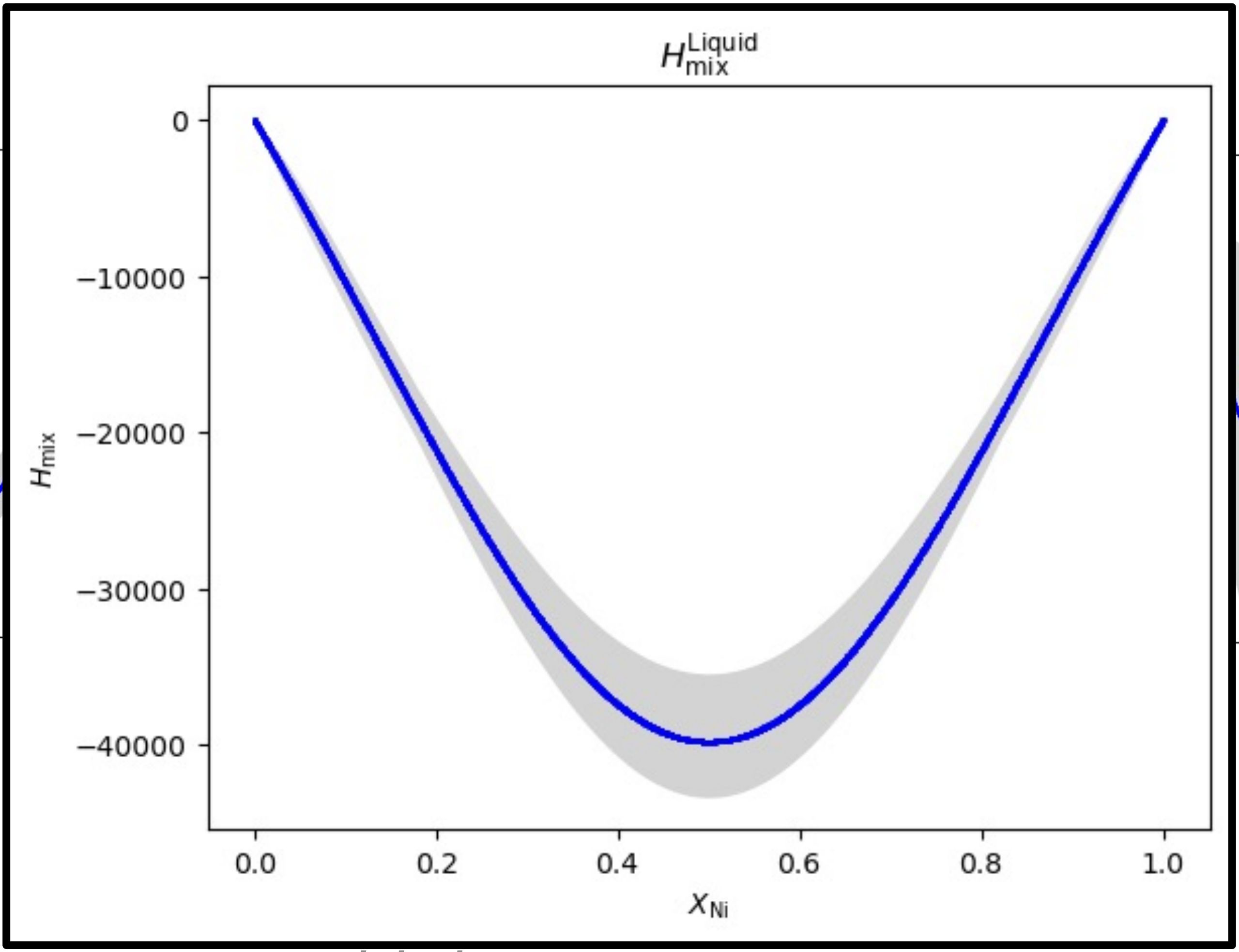


$$\sum_{i,j\neq i} x_i x_j \left(x_i - x_j\right)^{v}\ {}^{v}L$$

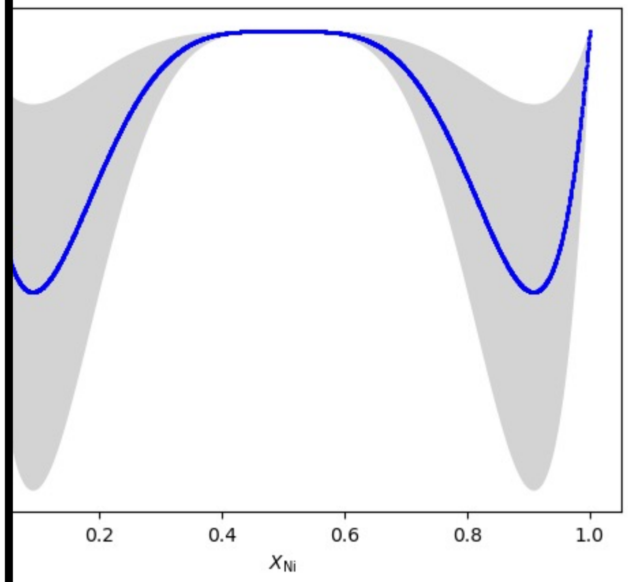
Uncertainty quantification in liquid interaction parameters

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



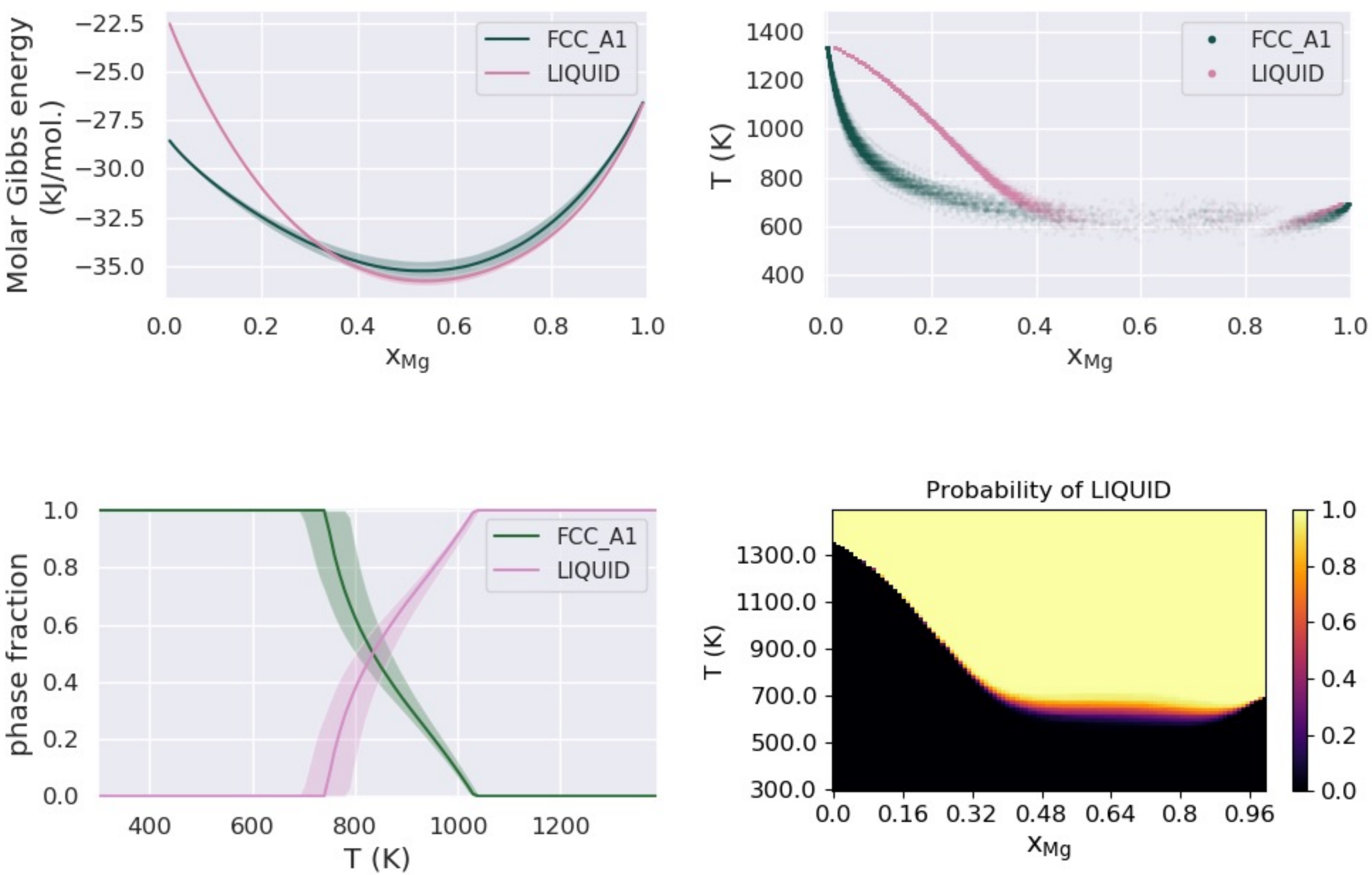


$^{2}L^{\mathrm{liquid}}$



$i,j\neq i$

Propagation of uncertainty to calculated properties and phase diagrams

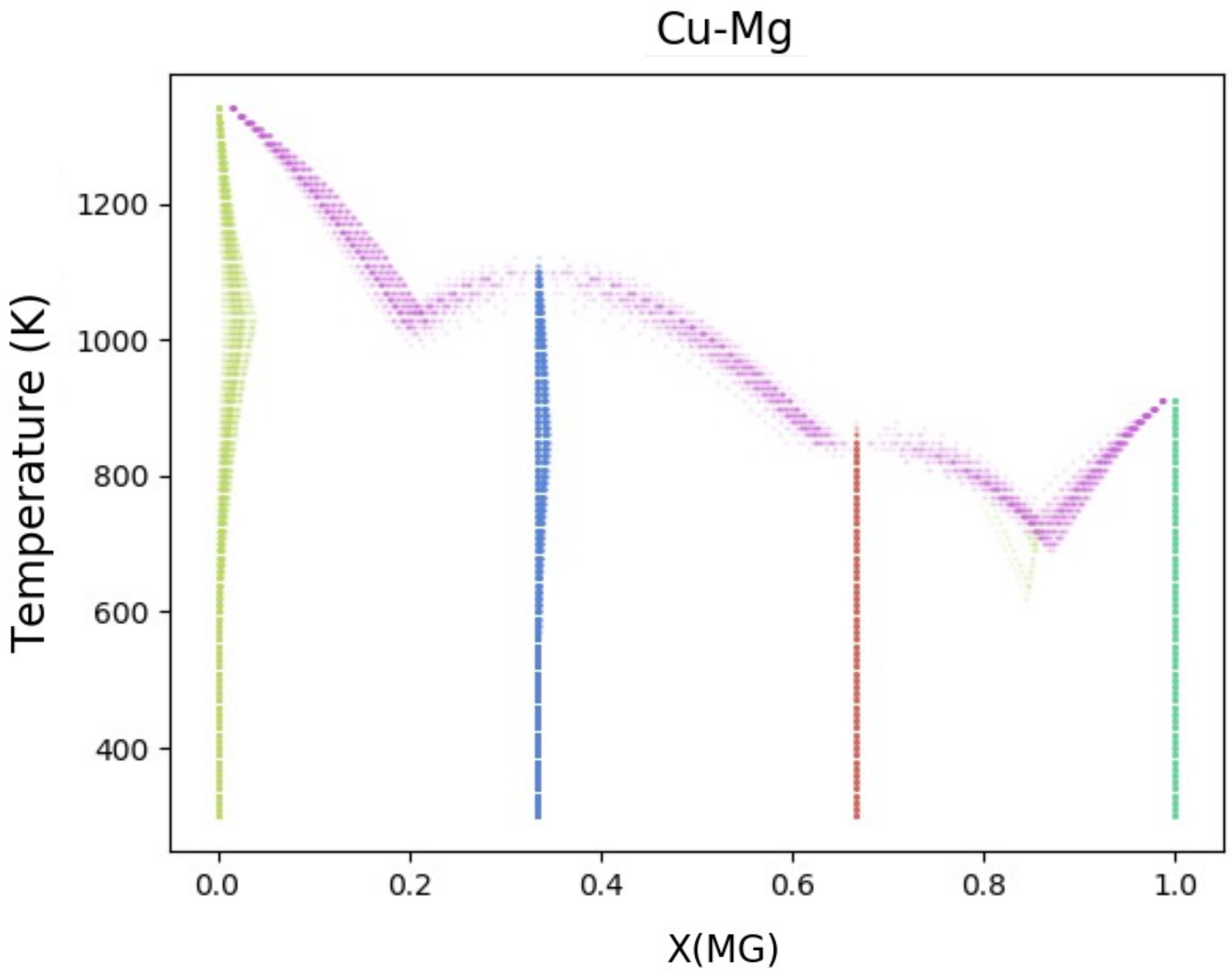


Paulson, Acta Mat. (2019)

Interactive Demo

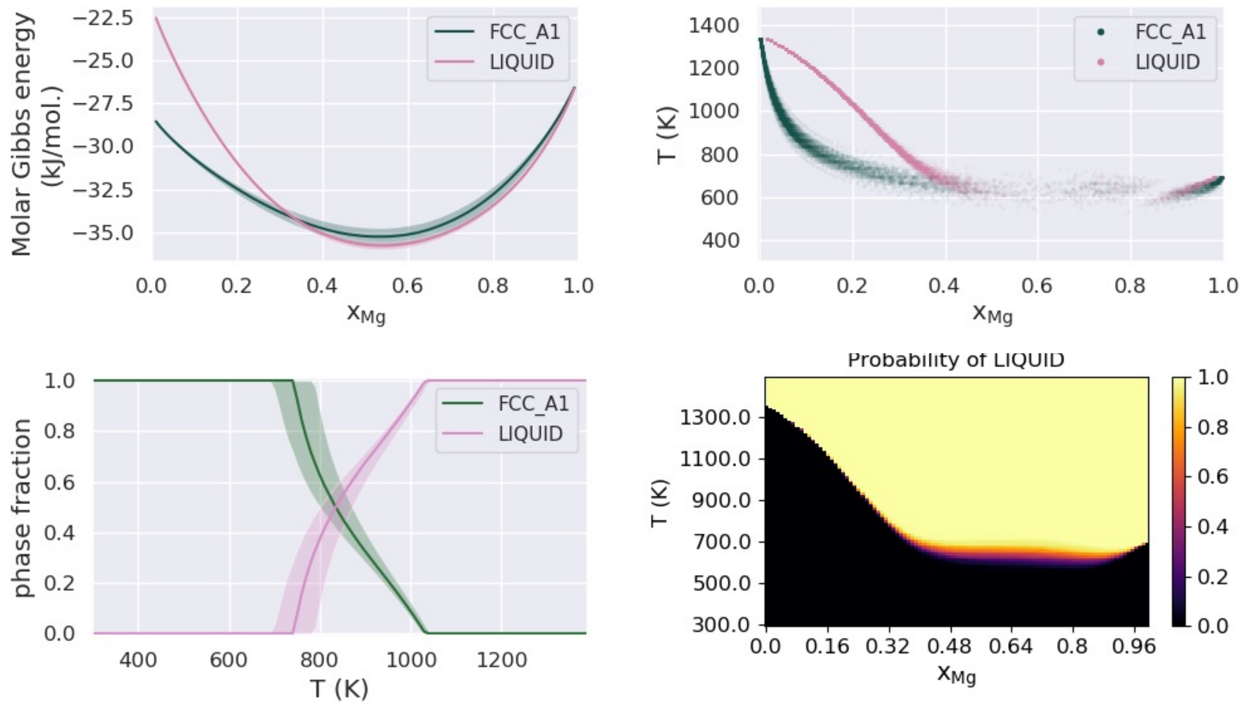
Uncertainty Propagation

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

Journal of MATERIALS RESEARCH

Invited Feature Paper

DOI: 10.1557/jmr.2020.269

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Sensitivity estimation for calculated phase equilibria

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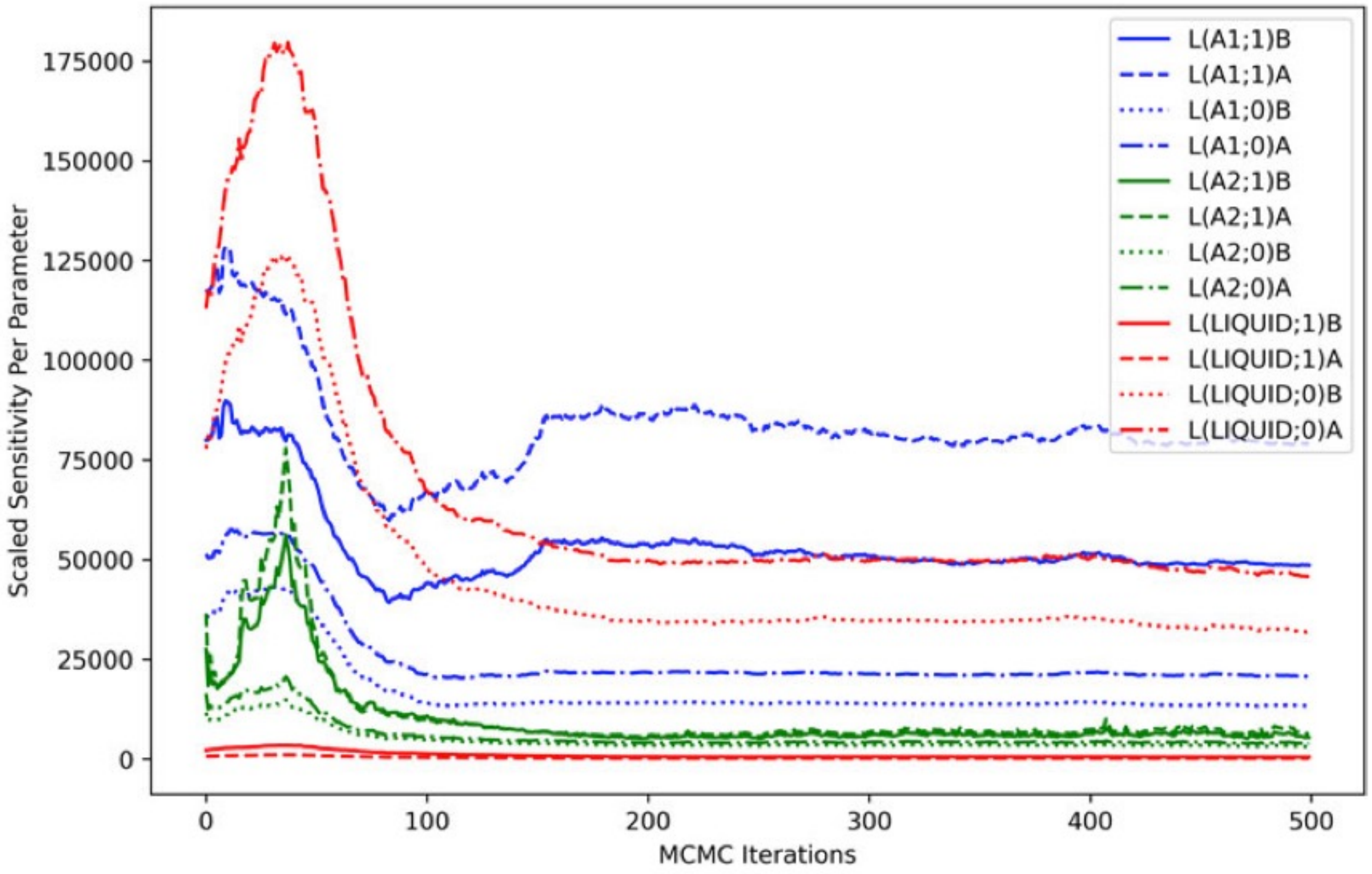
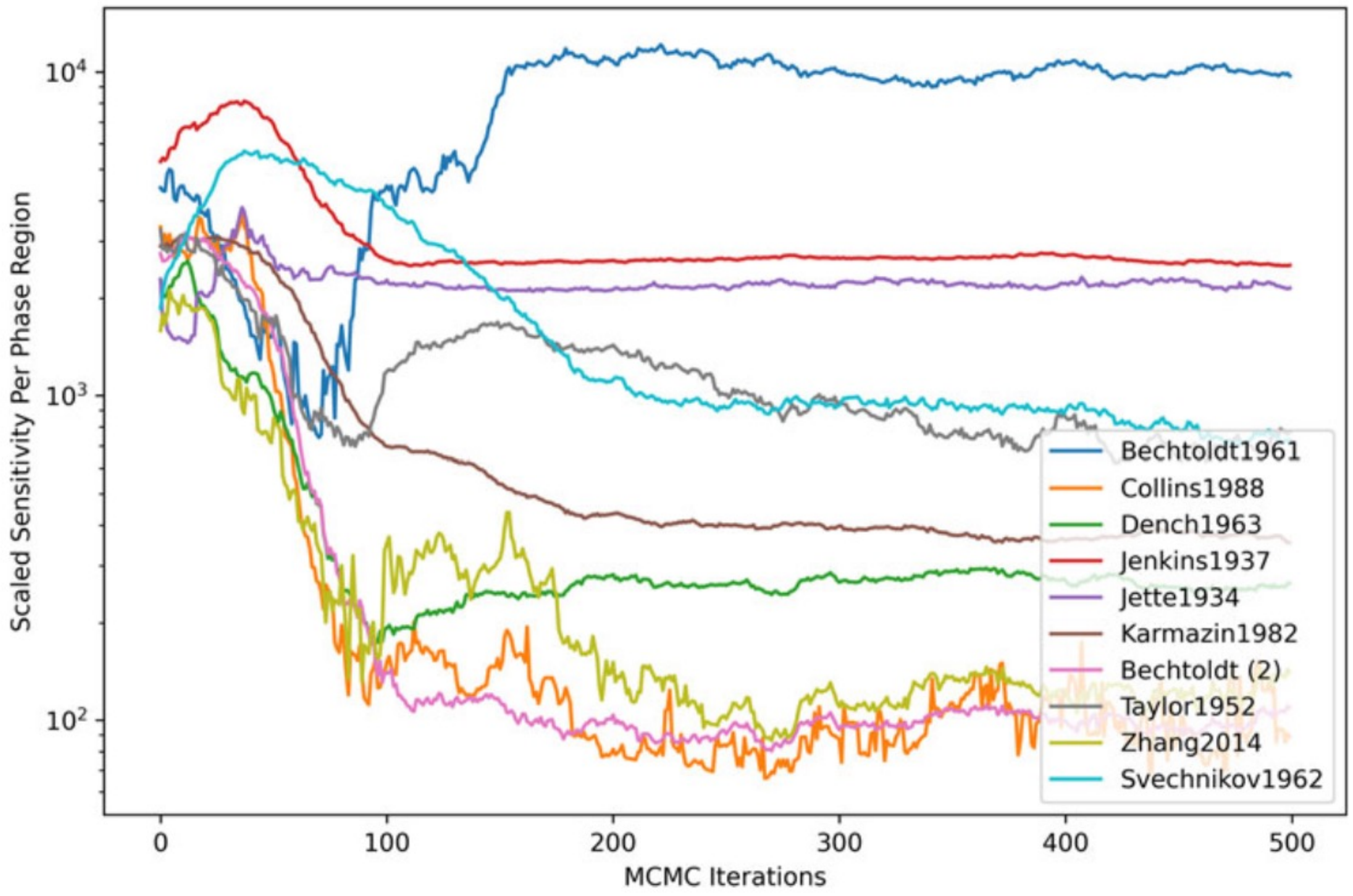
The development of a consistent framework for Calphad model sensitivity is necessary for the rational reduction of uncertainty via new models and experiments. In the present work, a sensitivity theory for Calphad was developed, and a closed-form expression for the log-likelihood gradient and Hessian 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



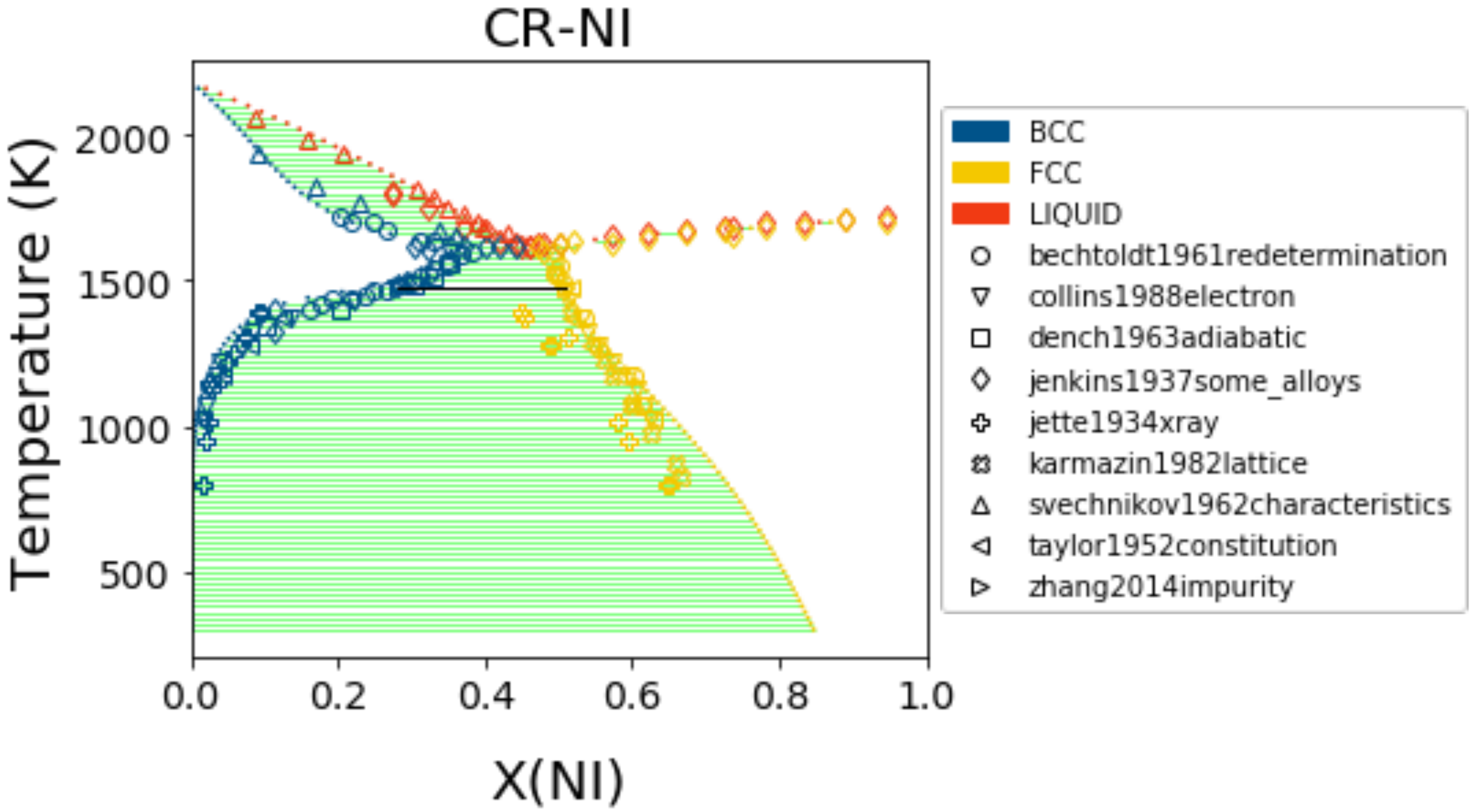
Otis, JMR (2020)

Cr-Ni Dataset Sensitivity



Otis, JMR (2020)

Cr-Ni Phase Diagram (2000 MCMC iterations)



Contact Us

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