

PRISMS Center

Center for PRedictive Integrated Structural Materials Science

CASM Tutorial – Intro

Brian Puchala, John C. Thomas, Anton Van der Ven

Anirudh Natarajan, John Goiri, Min-Hua Chen,
Max Radin, Jonathon Bechtel, Elizabeth Decolvenaere, Anna Belak,
Naga Sri Harsha Gunda, Sanjeev Kolli



TMS ICME 2017

Thurs. May 24, 2017

Website: prisms-center.org



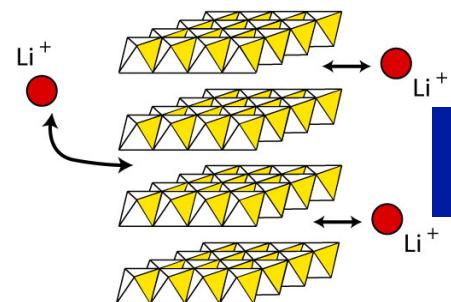
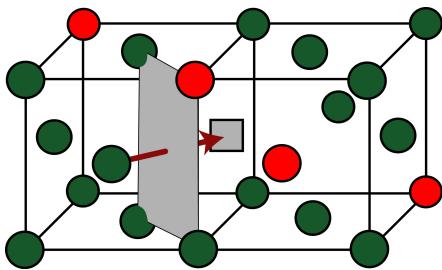
U.S. DEPARTMENT OF
ENERGY

Office of
Science

Center for PRedictive Integrated
Structural Materials Science

PRISMS

Electronic structure

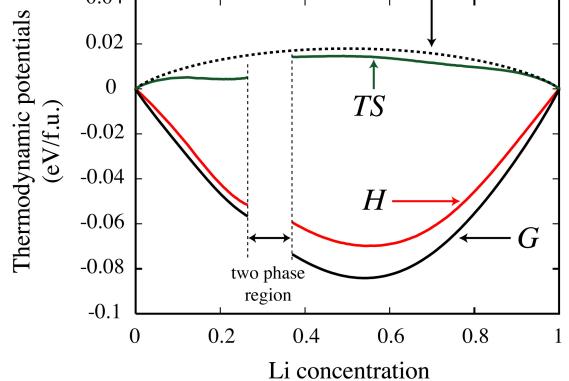


Statistical Mechanics

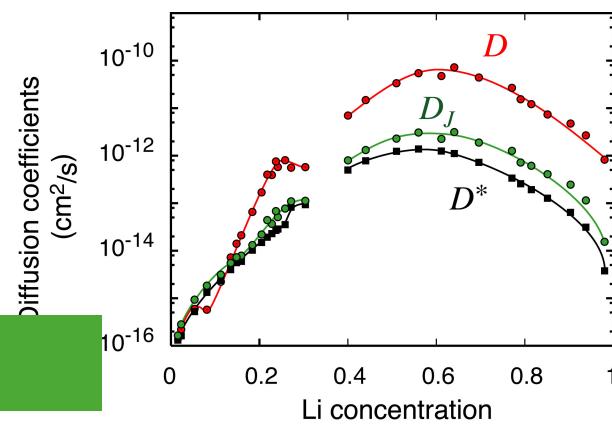
Thermodynamics

Kinetics

Solid Mechanics



Linking atomistic descriptions to phenomenological models of materials



Phenomenological descriptions

Electronic structure

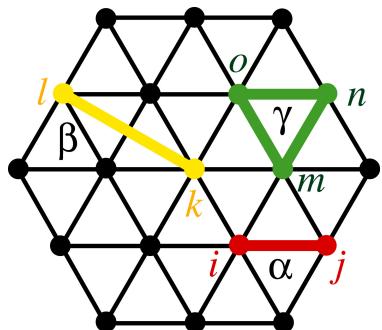
$$\left[-\nabla_i^2 + V_{ion}(r_i) + \int \frac{\rho(r')}{r - r'} dr' + V_{xc}(\rho(r_i)) \right] \phi_i(r_i) = \epsilon_i \phi_i(r_i)$$

Via rigorous and
arbitrarily extendible
cluster expansion
Hamiltonians

Cluster Expansion Hamiltonians

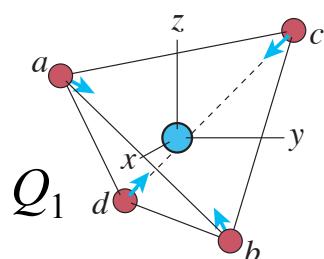
$$E(\vec{\sigma}) = V_o + \sum_i V_i \sigma_i + \sum_{i,j} V_{i,j} \sigma_i \sigma_j + \sum_{i,j,k} V_{i,j,k} \sigma_i \sigma_j \sigma_k + \dots$$

$$E(\vec{u}_1, \dots, \vec{u}_i, \dots, \vec{u}_N) = V_0 + \sum_{\alpha,m} V_m^{(\alpha)} \Phi_m^{(\alpha)}(\vec{Q}^{(\alpha)}) + \sum_{\beta,n} V_n^{(\beta)} \Phi_n^{(\beta)}(\vec{Q}^{(\beta)}) + \sum_{\gamma,p} V_p^{(\gamma)} \Phi_p^{(\gamma)}(\vec{Q}^{(\gamma)}) + \dots$$



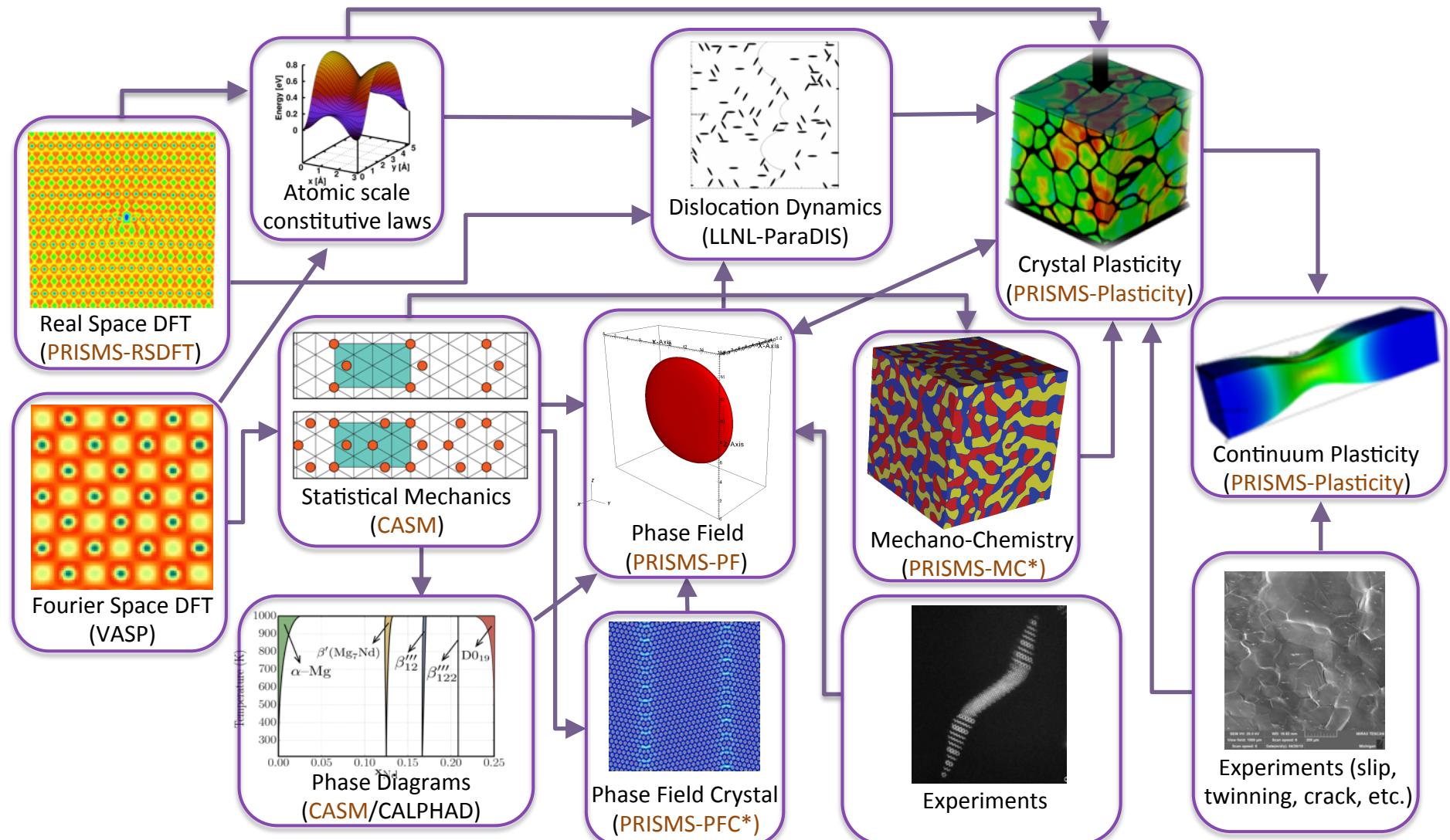
Statistical Mechanics

$$F = -k_B T \ln Z \quad Z = \sum_s \exp\left(-\frac{E_s}{k_B T}\right)$$



Phenomenological descriptions

PRISMS Center Integrated Framework



U.S. DEPARTMENT OF
ENERGY

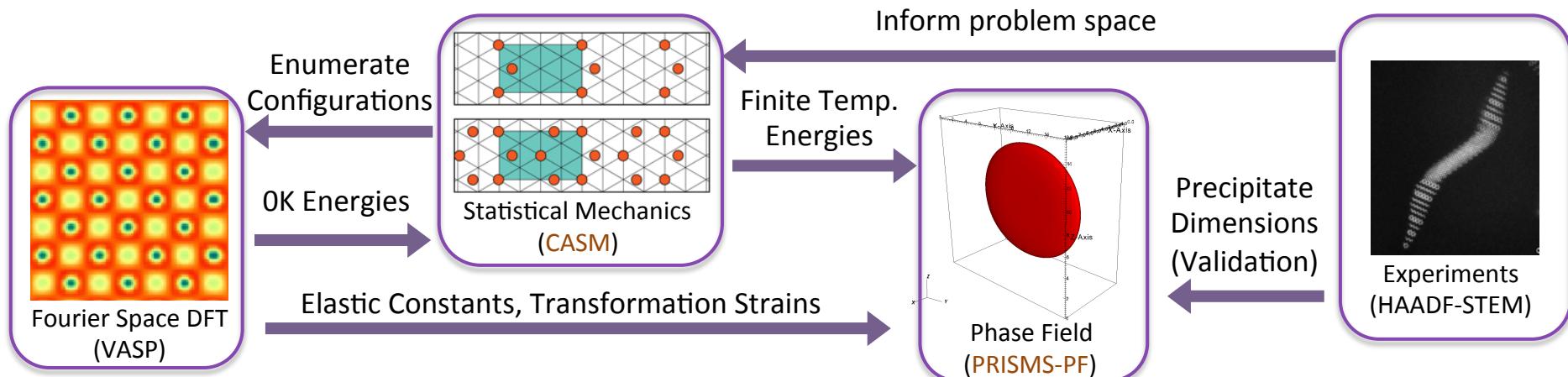
Office of
Science



Center for Predictive Integrated
Structural Materials Science

PRiSMS⁴

Using ICME to Investigate Mg-Nd β'' Composition and Morphology



Natarajan et al., Acta Mater. 2016
Natarajan and Van der Ven, Acta Mater. 2017



U.S. DEPARTMENT OF
ENERGY

Office of
Science

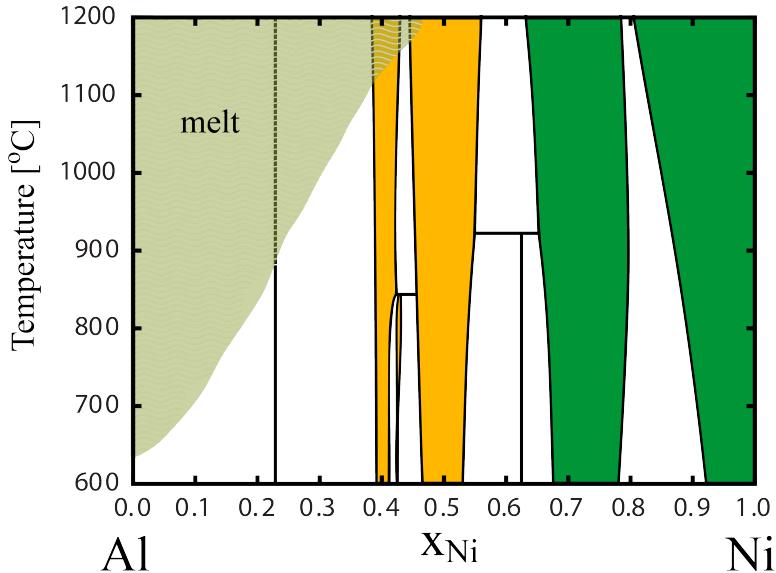


Center for Predictive Integrated
Structural Materials Science

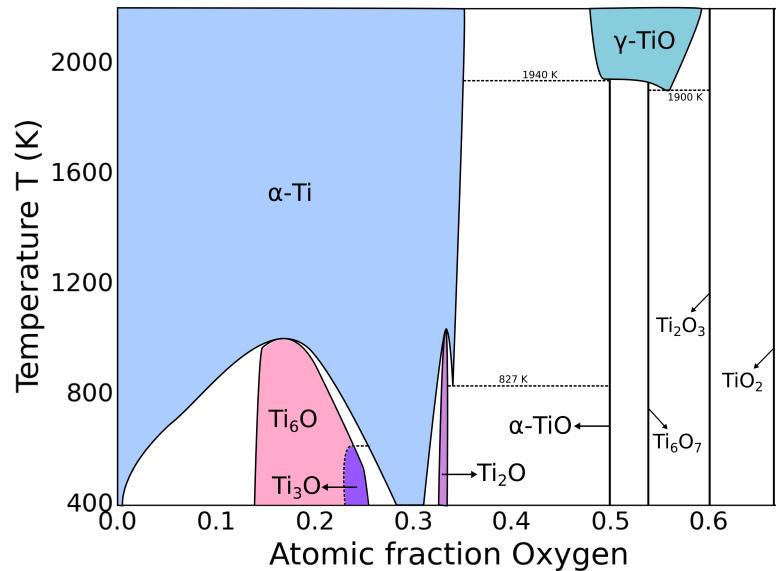
PRiSMS

Examples: Calculated phase diagrams

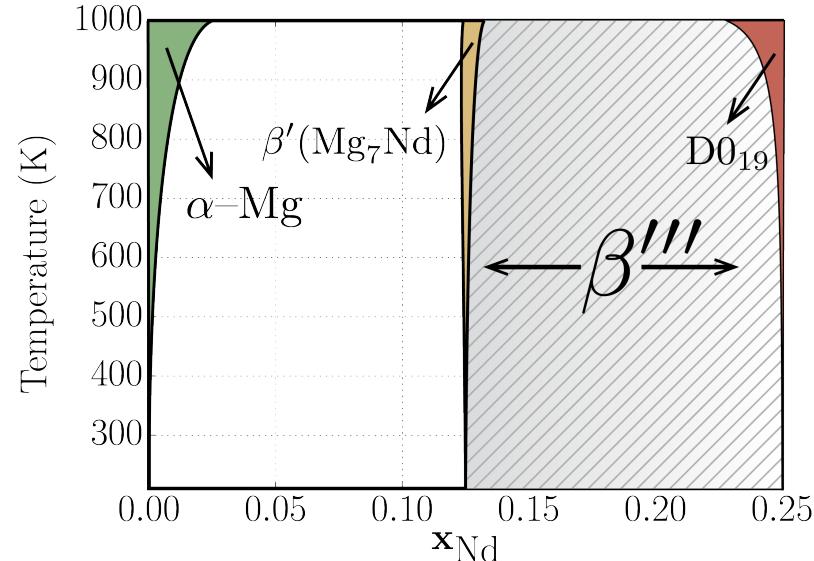
Ni-Al binary



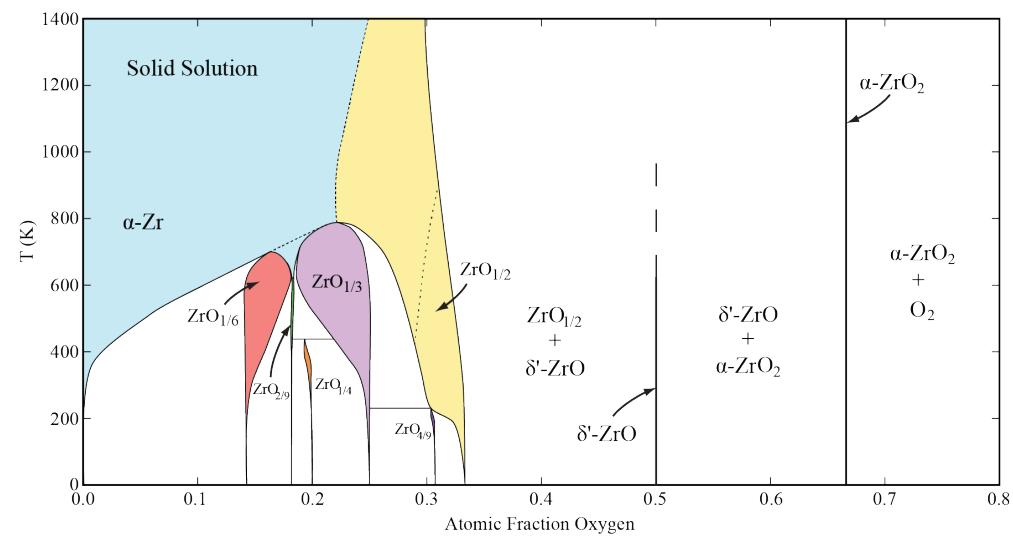
Ti-O binary



Mg-Nd binary



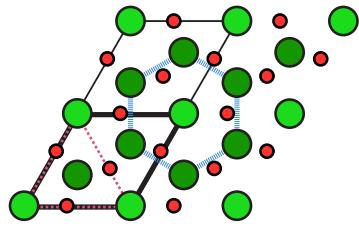
Zr-O binary



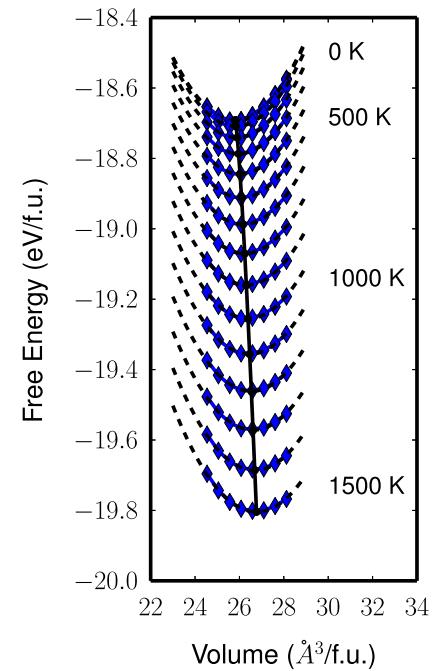
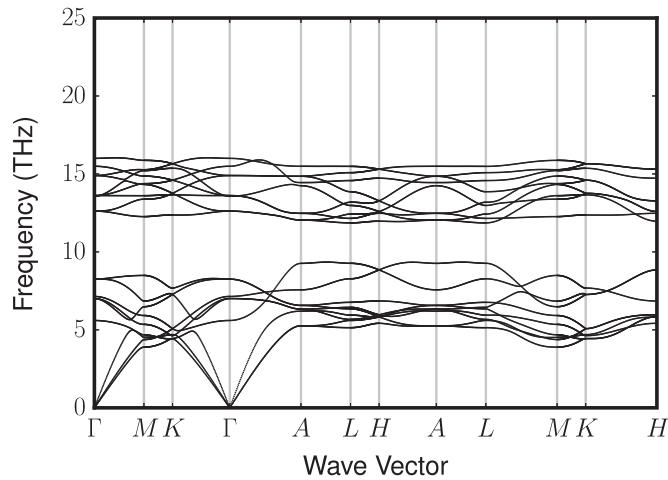
Examples: Vibrational excitations

Phonon Hamiltonian

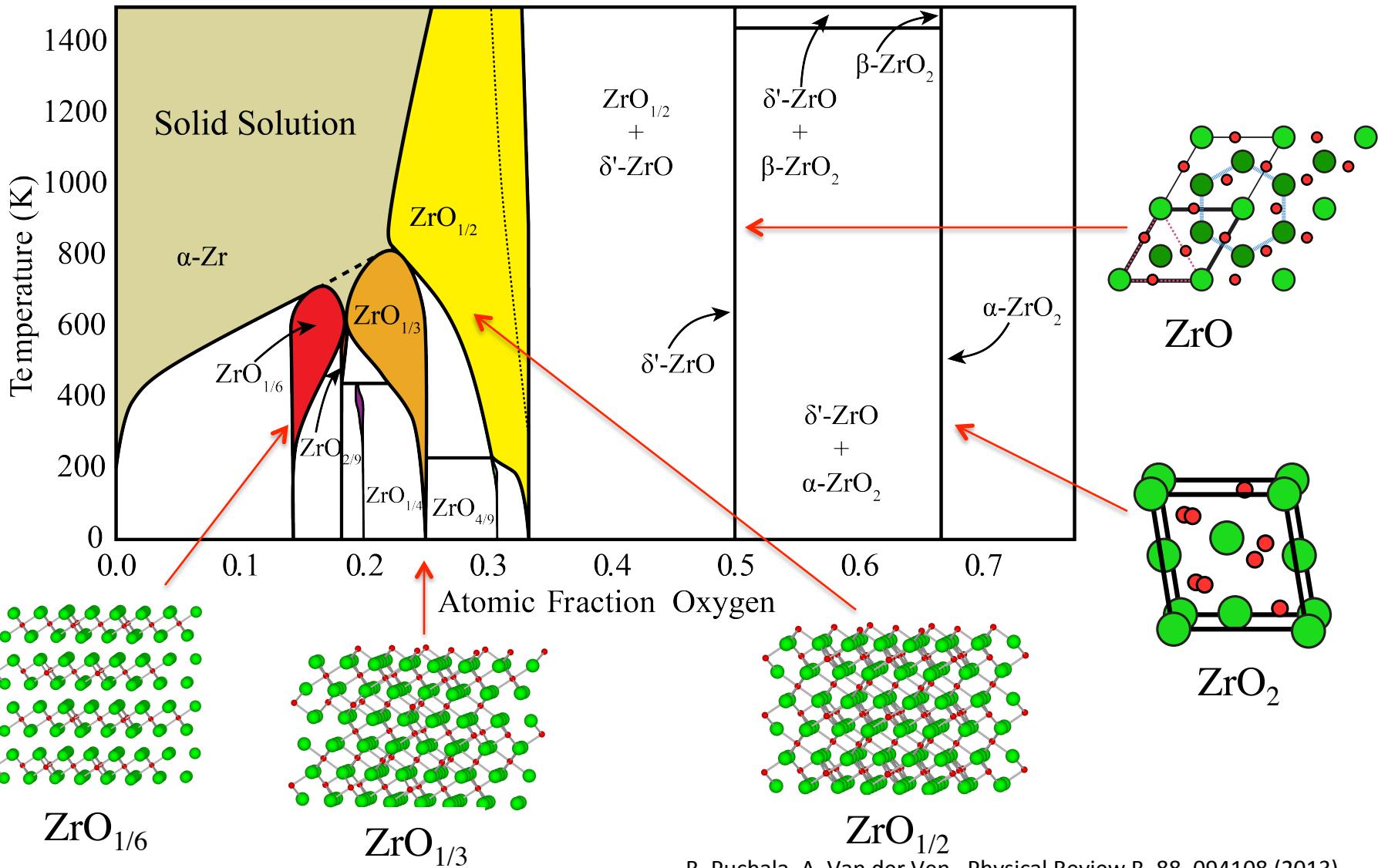
$$E(\vec{u}_1, \dots, \vec{u}_i, \dots, \vec{u}_N) = E_o + \frac{1}{2} \sum_{i,j} \sum_{\alpha,\beta} \Phi_{\alpha,\beta}^{i,j} \cdot u_{\alpha}^i \cdot u_{\beta}^j$$



ZrO



Examples: Calculated Zr-O phase diagram (configurational + vibrational excitations)

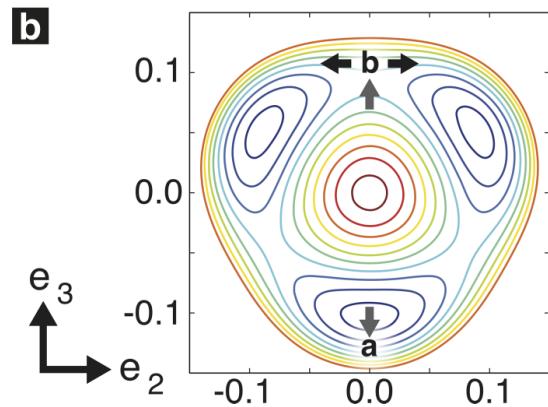
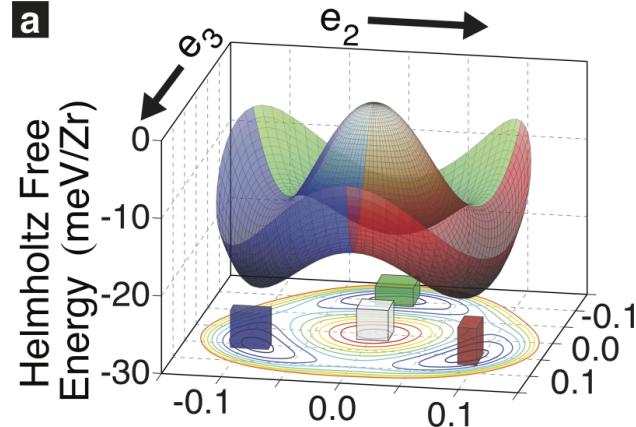


B. Puchala, A. Van der Ven, Physical Review B, 88, 094108 (2013).

M. H. Chen, B. Puchala, A. Van der Ven, CALPHAD (2015).

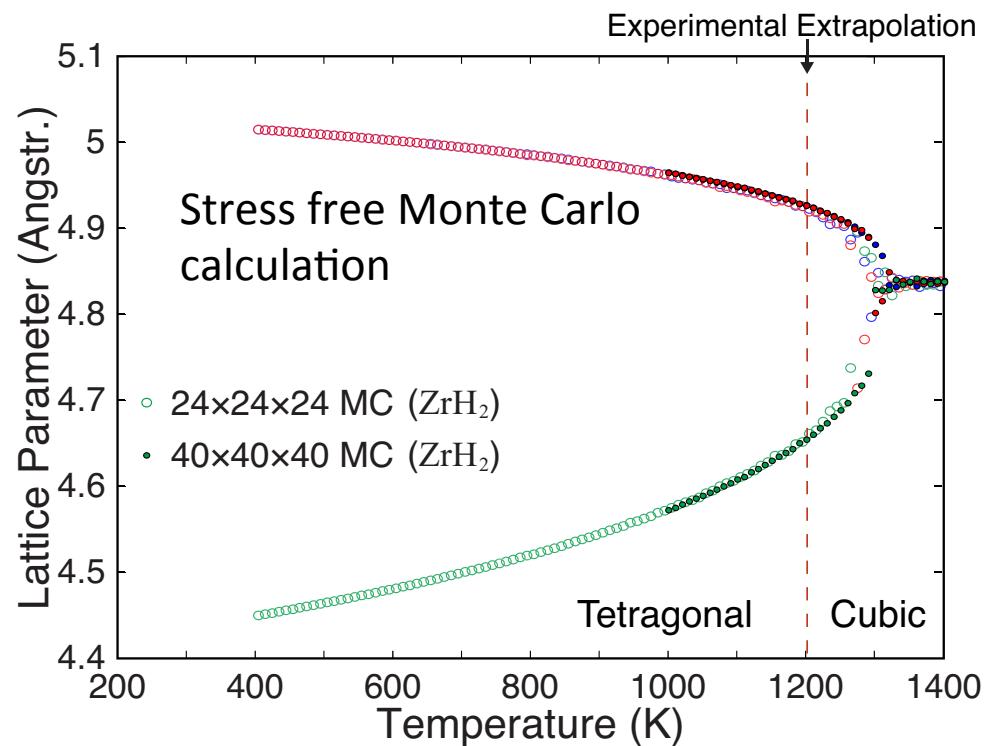
Examples: Anharmonic vibrational cluster expansion

Energy surface with homogeneous deviatoric deformation from DFT



$$E(\vec{u}_1, \dots, \vec{u}_i, \dots, \vec{u}_N) = V_0 + \sum_{\alpha,m} V_m^{(\alpha)} \Phi_m^{(\alpha)}(\vec{Q}^{(\alpha)}) + \sum_{\beta,n} V_n^{(\beta)} \Phi_n^{(\beta)}(\vec{Q}^{(\beta)}) + \sum_{\gamma,p} V_p^{(\gamma)} \Phi_p^{(\gamma)}(\vec{Q}^{(\gamma)}) + \dots$$

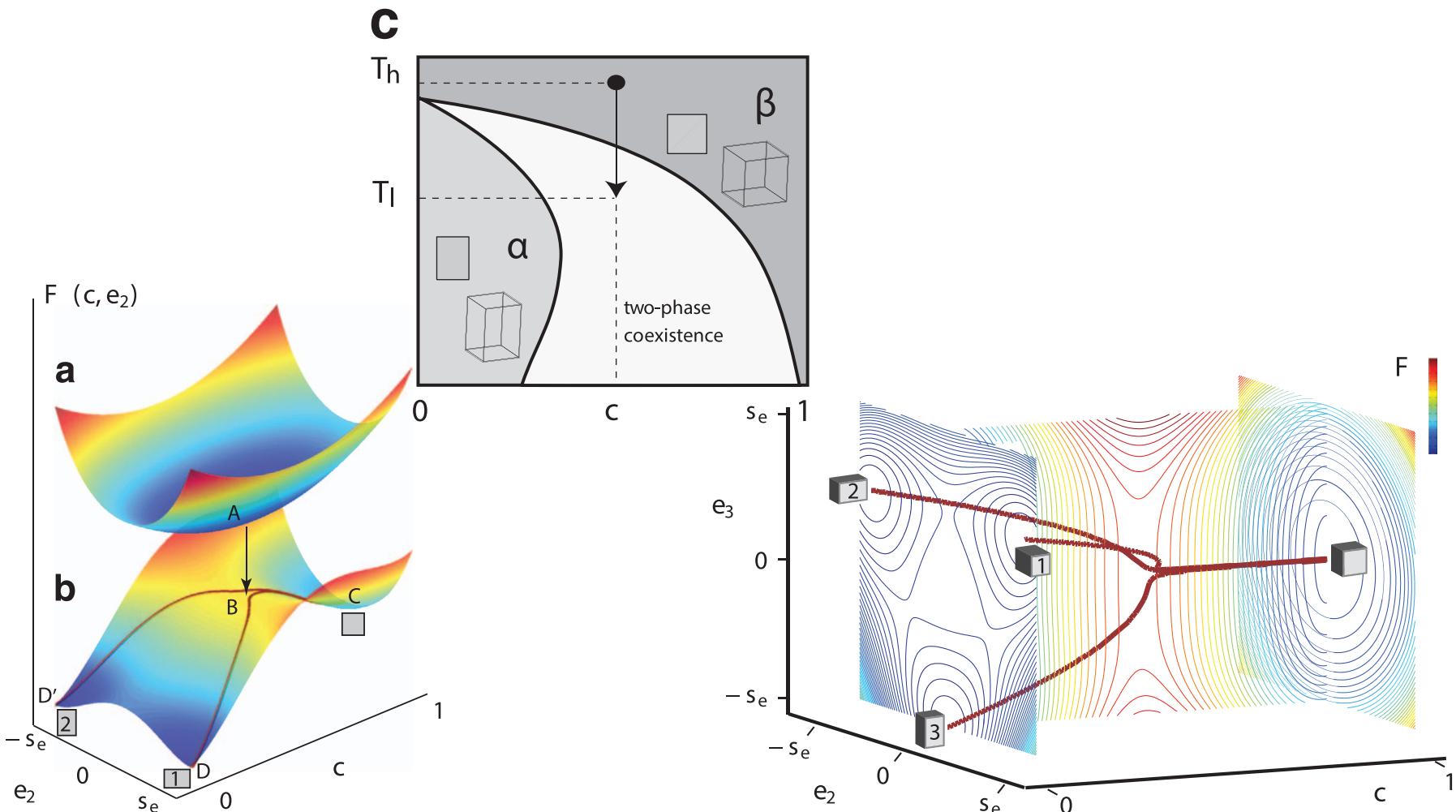
Cubic–tetragonal phase transition in ZrH_2



J.C. Thomas and A. Van der Ven, *Phys. Rev. B* **88**, 214111, (2013).

J.C. Thomas and A. Van der Ven, *Phys. Rev. B* **90**, 214105, (2014)

Examples: Chemo-mechanical free energy



Examples: Molecular occupation

Hybrid organic-inorganic ABX_3 perovskites: $\text{CH}_3\text{NH}_3\text{PbI}_3$

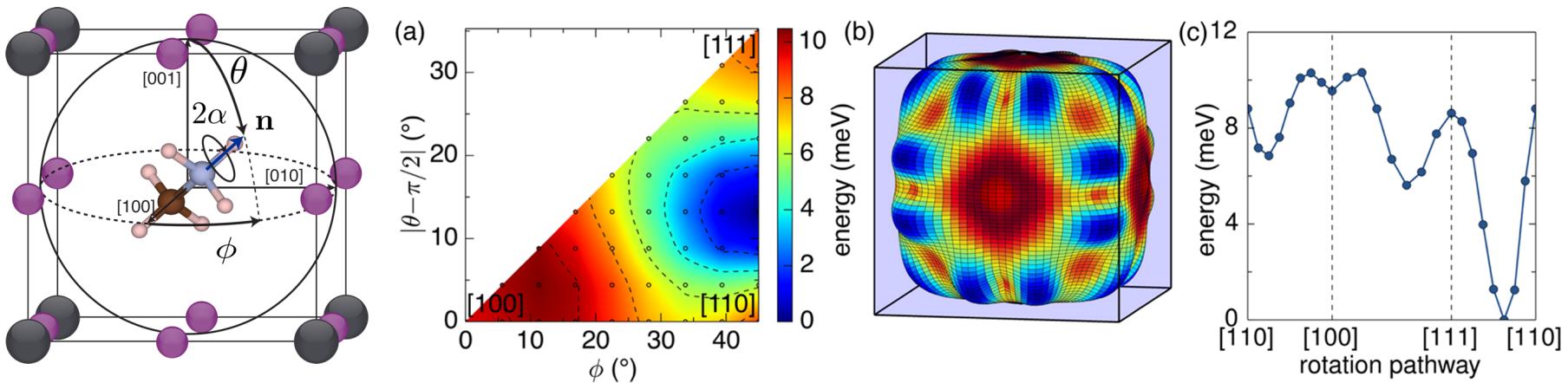


Figure 2. (a) Interpolated energy surfaces for molecular reorientation of CH_3NH_3^+ within cubic PbI_6 octahedral cages where the molecular geometrical center of mass resides at the A-site. At each orientation, the energy corresponding to the minimum energy on-axis rotation is plotted. (b) Polar plot of orientational energy surface where the radius is proportional to $|E - \beta E_{\max}|$ of scale from (a) where $\beta = 1 - 1/1000$. (c) Energies for selected rotational pathways along the edges of the asymmetric orientation region which represent rotations within the (001) and ($\bar{1}10$) lattice planes via $[100] \rightarrow [110]$ and $[001] \rightarrow [111] \rightarrow [110]$ rotations, respectively.

Examples: Molecular occupation

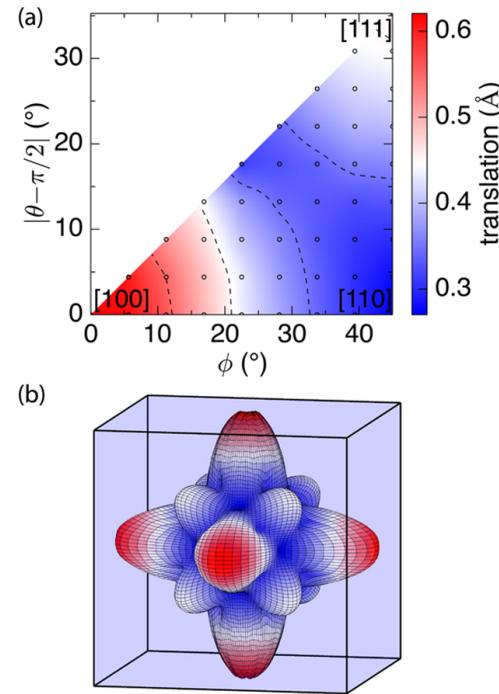
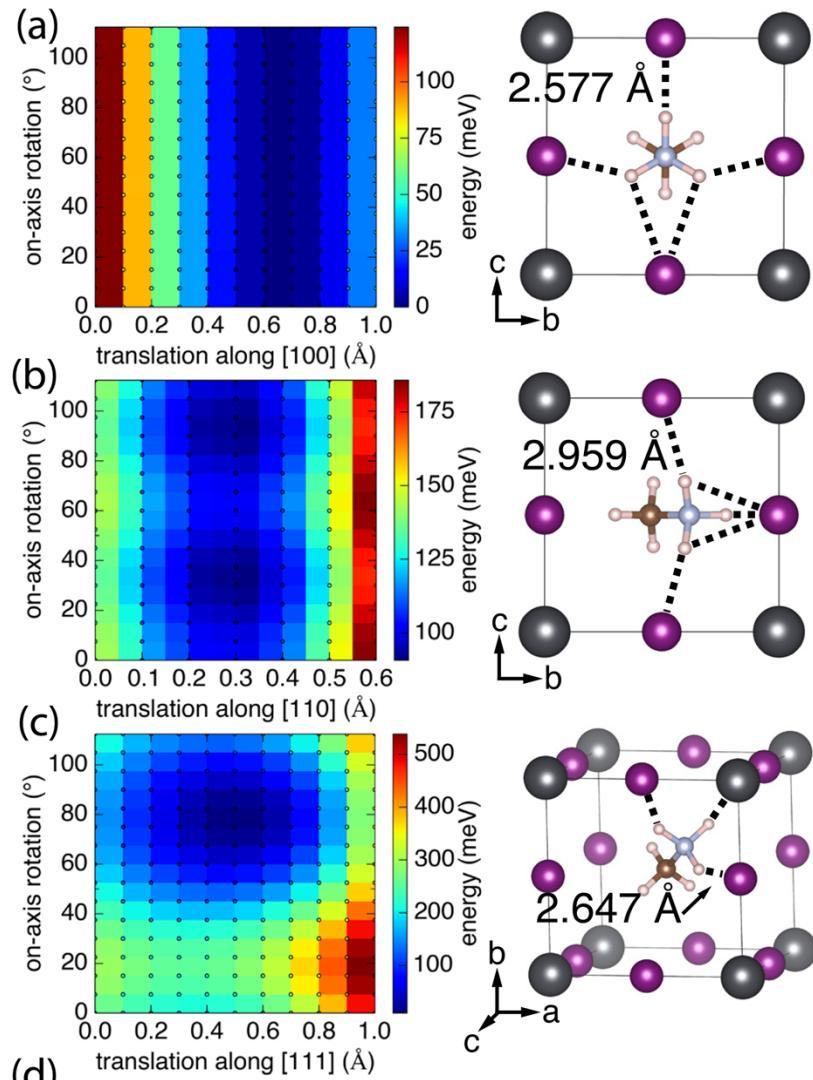


Figure 4. (a) Within the asymmetric orientation region, the translations associated with the lowest energy configurations are plotted. The inclination and azimuthal angles are given relative to the [100] reference configuration. (b) Polar plot where the radius is proportional to the minimum energy translation providing a representation of the locus of minimum energy molecular translations throughout the cubic unit cell.

CASM Workflow

- Problem specification
 - Primitive crystal structure and allowed degrees of freedom
- Enumerate configurations
- Electronic structure calculations:
 - Specify calculation parameters
- Effective Hamiltonian specification:
 - Specify cluster expansion basis set
- Fit coefficients
- Evaluate effective Hamiltonian:
 - Monte Carlo calculation of ensemble averages
 - Evaluate strain polynomials
- Fit phenomenological models
 - Free energy, mobilities
 - For analysis and as input to models at greater time/length scales

CASM Design Principles

Flexibility

- CASM provides a set of tools and enables customization at many steps
- As much as possible, CASM does not force you to use a particular method for enumeration, calculation, model fitting, etc.

Reproducibility

- Log all commands over the course of a project
- Try to record provenance of all data as part of normal workflow

Data Management and Automation

- Tools to generate, screen, and dispatch DFT calculations, and then organize, filter, and analyze the results.
- Managed directory tree and dependency framework that allows multiple calculation and model parameters to be tested and compared within a single project

CASM Design Principles

Interoperability

- Most input and output files are stored in JavaScript Object Notation (JSON) file formats, which is widely used and highly readable.
- Weak coupling between CASM and DFT codes, so that CASM can be adapted to work with various existing and future codes.

Collaboration

- Easy archival of projects ('casm files' command), with granular control over amount and type of data to include.
- Parameterized physics models can be shared without DFT training data and used to perform finite-temperature calculations

Efficiency

- CASM implements unique and highly optimized algorithms for generating and analyzing data and building physics models.
- CASM generates optimized C++ code to evaluate basis functions which is automatically compiled and used in the Monte Carlo code, enabling larger simulations and improved convergence.

CASM Components:

- "casm" command line executable, with JSON input files
 - C++11 for "core" (symmetry, crystallography, basis sets, Monte Carlo)
 - Doxygen documentation
- Python wrappers, visualization, analysis
 - "casm" Python module for standard usage interfaces
 - DFT software wrappers for VASP & SeqQuest
 - "casm-learn" coefficient fitting integrates with scikit-learn, deap (genetic algorithms)
 - Easy integration with VESTA for viewing configurations
 - Interactive visualization
- Python PBS & Slurm interface for cluster job submission:

CASM Components:

- Public version:
 - Occupation DoF & grand canonical Monte Carlo calculations
 - "beta" canonical Monte Carlo calculations enabling a large number of possible occupation constraints
 - Variance-constrained Monte Carlo to be released very soon
 - Available on github:
 - <https://github.com/prisms-center/CASMcode>
 - Demo project with pre-calculated DFT training data:
 - https://github.com/prisms-center/CASMcode_demo

Near Future (next 0-2 yrs):

Much of this is also completed in part or in full internally, and needs to be completed or polished in some way for the public code.

- Kinetics:
 - Enumeration of atomic hops and local configurations, NEB calculation, local cluster expansion of activation energy, KMC
- Phonons:
 - Enumerate configurations for force constant calculation, dynamical matrix calculation, free energy calculation in quasi-harmonic approximation
- Strain polynomials:
 - Generate symmetrized strain Hamiltonian, enumerate strained configurations, DFT calculation, fit Hamiltonian coefficients
- Gamma-surface:
 - Setup gamma-surface geometries and perform DFT calculations
- Composition & configurational order parameter dependent free energies
 - Specify ordered configurations, generate order parameter, perform variance-constrained Monte Carlo calculations and free energy integration
- Machine learning tools for optimal design of Monte Carlo calculations
- Integration with Materials Commons



This repository

Search

Pull requests Issues Gist



prisms-center / CASMcode

Unwatch ▾ 15

★ Unstar 5

Fork 4

Code

Issues 0

Pull requests 0

Wiki

Pulse

Graphs

Settings

First-principles statistical mechanical software for the study of multi-component crystalline solids — [Edit](#)

3 commits

2 branches

1 release

2 contributors

Branch: master ▾

New pull request

New file

Upload files

Find file

SSH ▾

git@github.com:prisms-cente



Download ZIP

README.md

v0.2.1
Now Available!

CASM: A Clusters Approach to Statistical Mechanics

CASM (<https://github.com/prisms-center/CASMcode>) is an open source software package designed to perform first-principles statistical mechanical studies of multi-component crystalline solids. CASM interfaces with first-principles electronic structure codes, automates the construction and parameterization of effective Hamiltonians and subsequently builds highly optimized (kinetic) Monte Carlo codes to predict finite-temperature thermodynamic and kinetic properties. CASM uses group theoretic techniques that take full advantage of crystal symmetry in order to rigorously construct effective Hamiltonians for almost arbitrary degrees of freedom in crystalline solids. This includes cluster expansions for configurational disorder in multi-component solids and lattice-dynamical effective Hamiltonians for vibrational degrees of freedom involved in structural phase transitions.

