DOE CSGF Review 2023

July 18th 2023

The foundation for a ground-up & robust approach to computational magnetic materials discovery

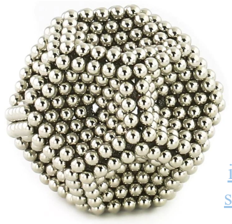
Presenter: Guy Moore

Mentor: Dr. Matthew Horton
Mentor: Dr. Sinéad Griffin
Practicum mentor: Dr. Tae Wook Heo
Advisor & Mentor: Dr. Kristin Persson

Understanding magnetic properties - essential for many applications

Permanent magnets

Alternatives to rare earths



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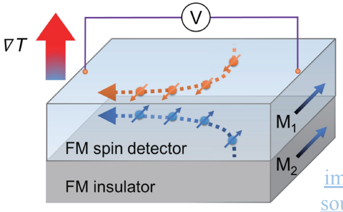
Spintronics

Spin Seebeck effect depicted



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Magnetic cooling

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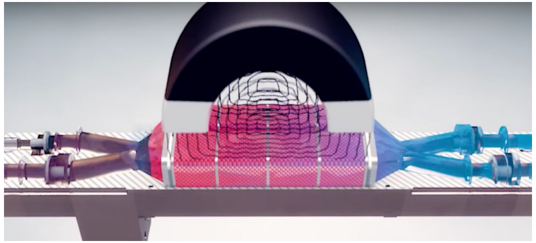


We address several limitations in computational predictions of magnetic properties

Challenges

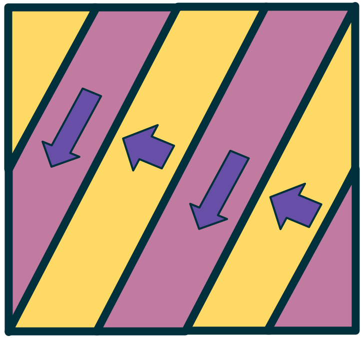
- Noncollinear ground-state → Many properties
- Ground-state → high-dimensional optimization & computational cost
- Extensions to finite-temperature & microstructure effects?

Solutions

- Unified ground-up computational framework
- Robust computation of many magnetic properties from first principles

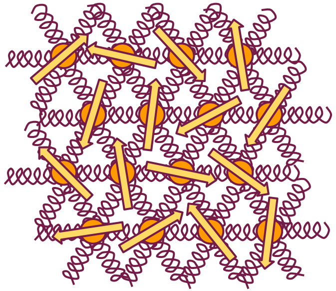
Outline of this talk: Multiscale magnetic framework

Part IV — **Continuum**
Phase field + Micromagnetic



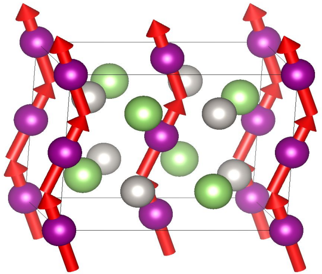
↑ Hubbard-Stratonovich transformation

Part III — **Lattice models**
Monte Carlo



↑ Green's function to tight-binding

Part II
Part I — **Atomistic**
Density functional theory



MnPtGa

Part I:

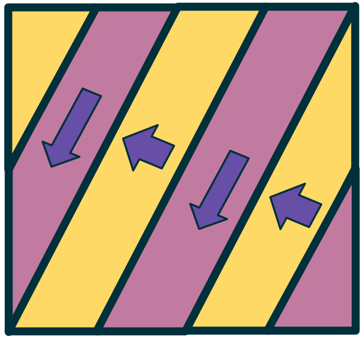
Linear response DFT+U+J

&

Source-free B_{xc}

Multiscale magnetic framework

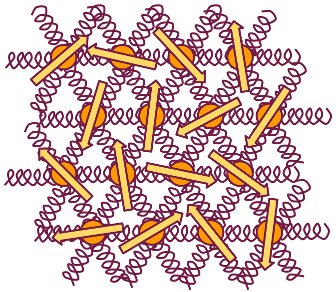
Continuum
Phase field +
Micromagnetic



Hubbard-Stratonovich transformation

Part III

Lattice models
Monte Carlo

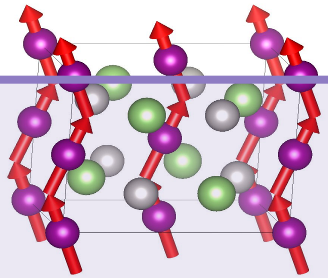


Green's function to tight-binding

Part II

Part I

Atomistic
Density functional theory

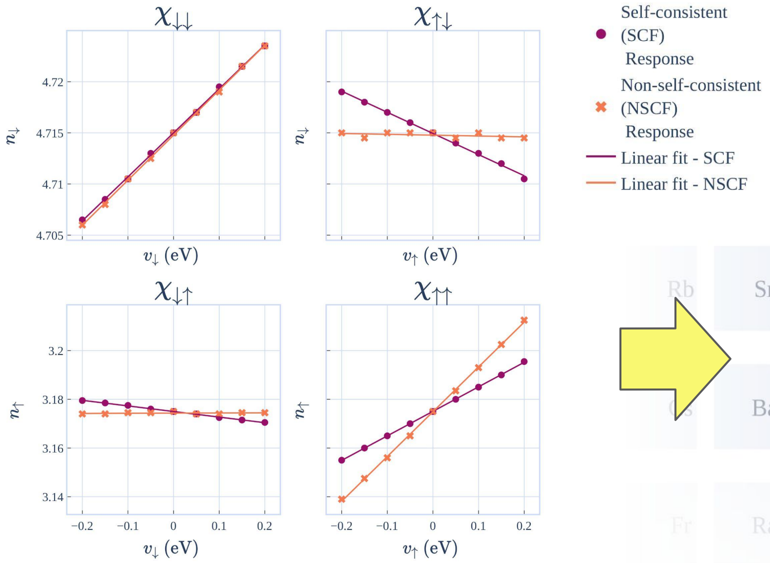


MnPtGa

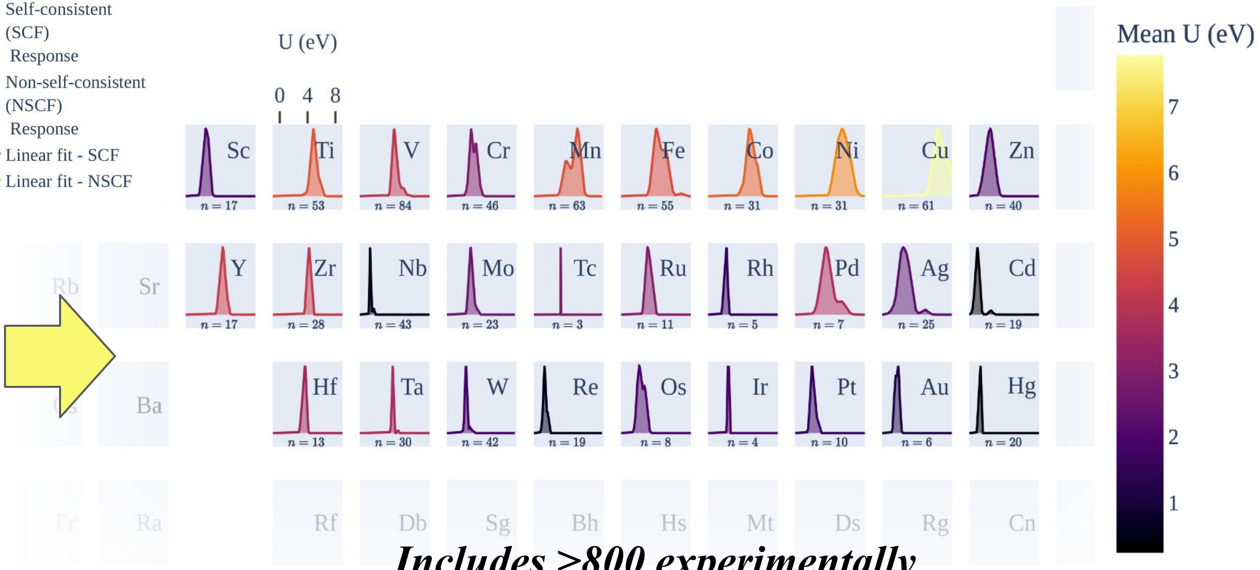
Identifying exotic magnetic ground-states in a robust manner

Linear response U & J workflow (`atomate`)

Spin-polarized linear response (Ni-d states in NiO)



Hubbard U and Hund J values



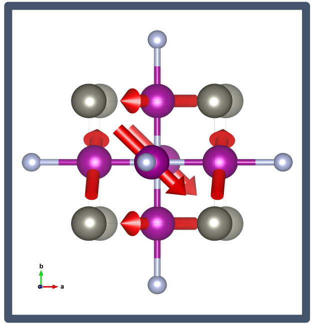
Includes >800 experimentally synthesizable materials

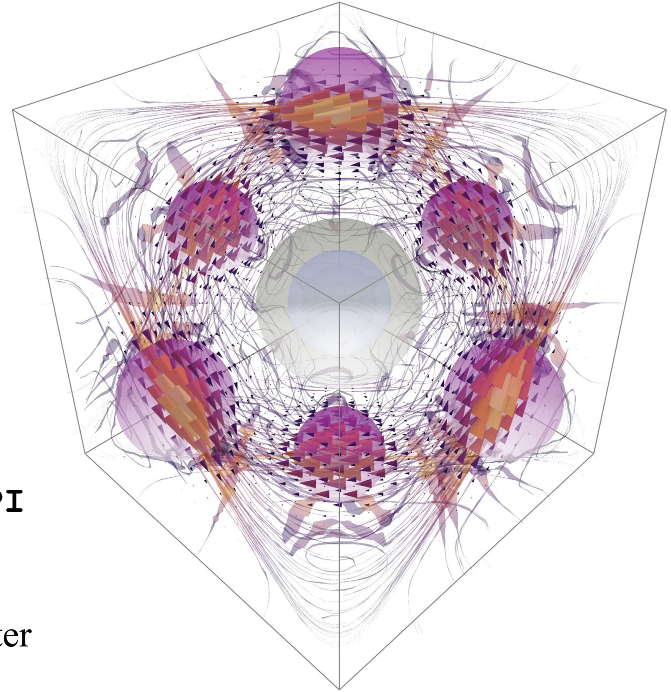
Preprint: *arXiv:2201.04213*

Source-free XC functional

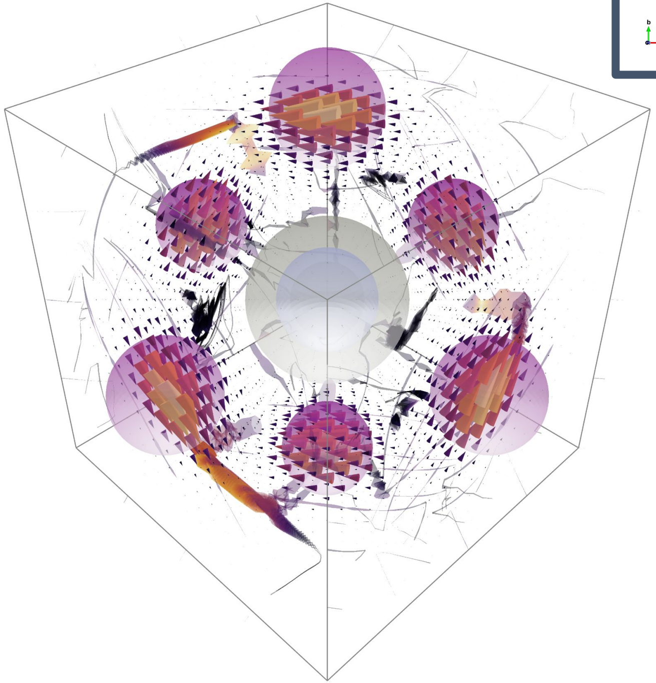
Test case: Mn_3ZnN

Based on the theoretical work of Sharma et. al. (DOI: 10.1021/acs.jctc.7b01049)





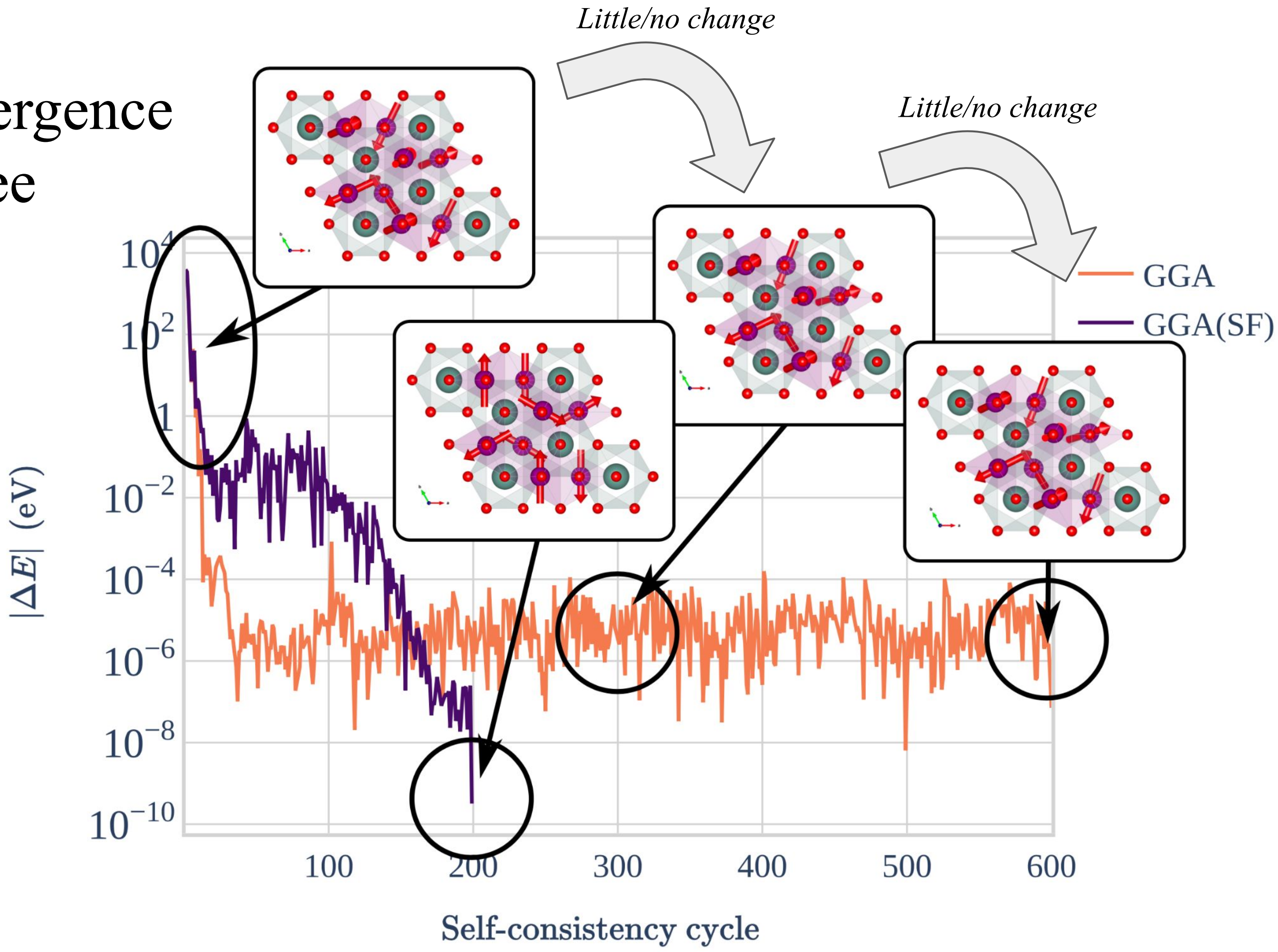
Source-free GGA+U+J



Conventional GGA+U+J (SDFT)

`OpenACC` + **`MPI`**
parallelized
&
Run on Perlmutter
GPU nodes

Improved convergence using source-free functional



Case study: $YMnO_3$

Part II:
`SpinPSO` *noncollinear ground-state optimization*

`SpinPSO` Algorithm

Identifying noncollinear magnetic ground-states

Stochastic agent-based algorithm based on the combined dynamical frameworks of:

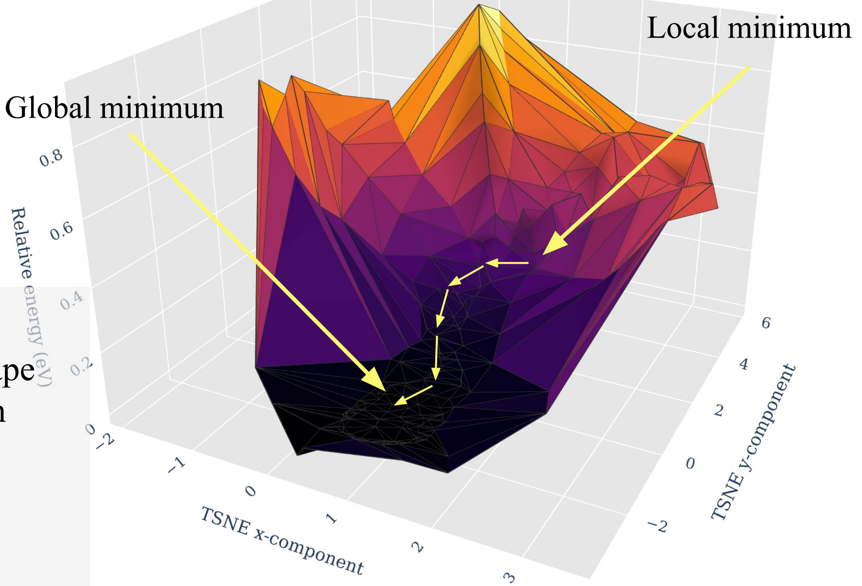
- Atomistic spin dynamics
- Particle swarm optimization (PSO)

Inspired by previous approaches NEB (Fidimag) and Firefly algo. (PyChemia)

$$\mathbf{h}_{i,j}^{n} = f\left(\tilde{\mathbf{s}}_{i,j}^{n}, \hat{\mathbf{s}}_{j}^{n}, \left(\mathbf{h}_{\text{eff}}\right)_{i,j}^{n}\right)$$

agent best — swarm best — local fields *from constrained moment DFT*

$$\frac{\Delta \mathbf{s}_{i,j}}{\Delta \tau} = \frac{\mathbf{s}_{i,j}^{n+1} - \mathbf{s}_{i,j}^{n}}{\Delta \tau} = -\lambda \mathbf{s}_{i,j}^{n} \times \mathbf{h}_{i,j}^{n} - \alpha \mathbf{s}_{i,j}^{n} \times \left(\mathbf{s}_{i,j}^{n} \times \mathbf{h}_{i,j}^{n}\right)$$



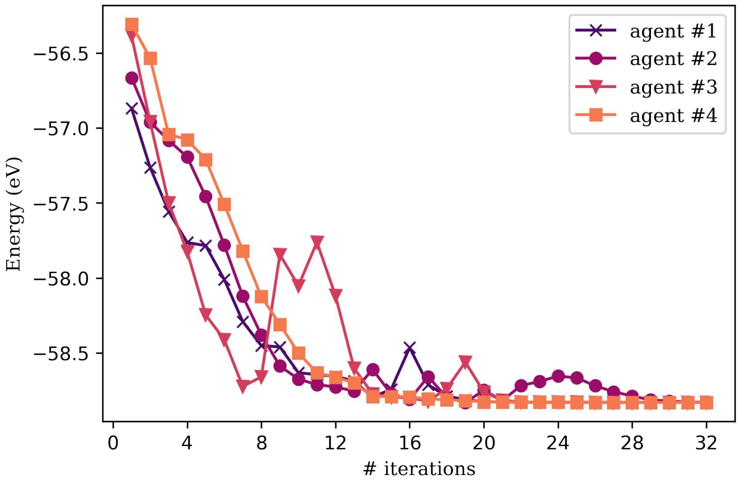
Reduced-dimension potential energy landscape for FeF_3 spin orientation

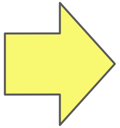
Energies from PBE

Visualized using t-distributed stochastic neighbor embedding (t-SNE)

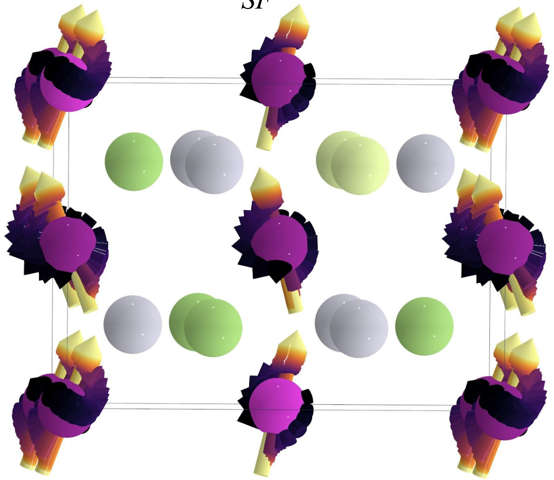
`SpinPSO` algorithm for noncollinear magnetic ground-states

Test case: MnPtGa





*Computed ground-state configuration using **`SpinPSO`** + GGA_{SF}+U+J*



MnPtGa

Visualized using Crystal Toolkit

Part III:

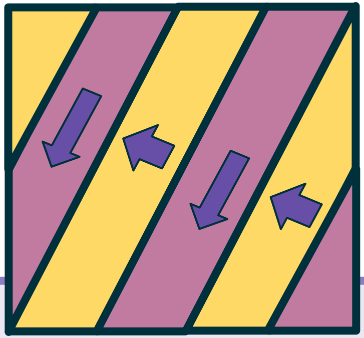
Spin-lattice Hamiltonians

&

Monte Carlo method to probe the thermodynamics

Multiscale magnetic framework

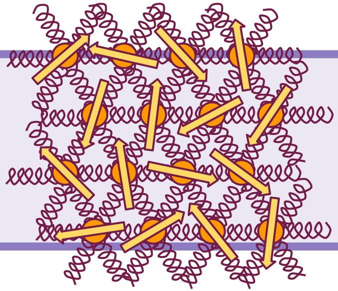
Continuum
Phase field +
Micromagnetic



Hubbard-Stratonovich transformation

Part III

Lattice models
Monte Carlo

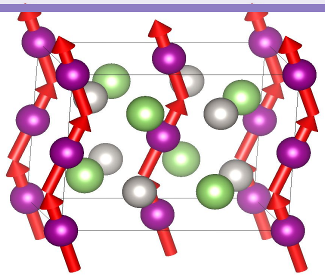


Green's function to tight-binding

Part II

Part I

Atomistic
Density functional theory

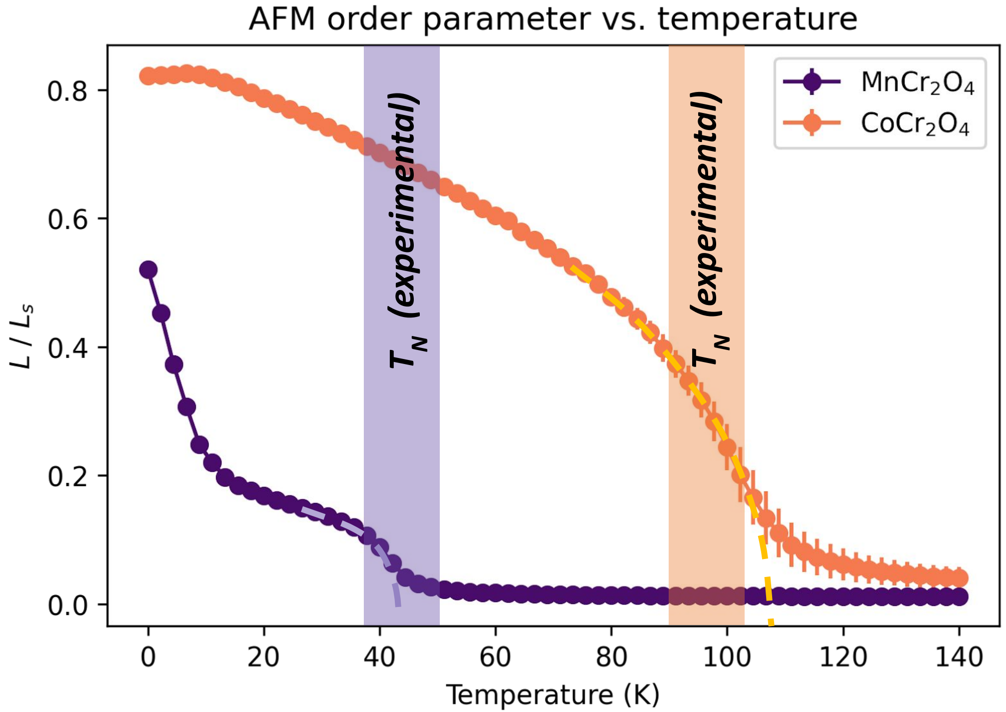


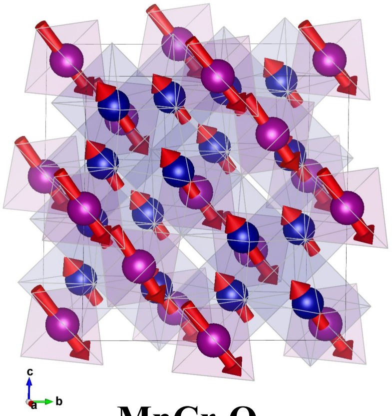
MnPtGa

Using custom **Cython** Monte Carlo code

MPI+**OpenMP** parallelized

Successful prediction of experimental transition temperatures





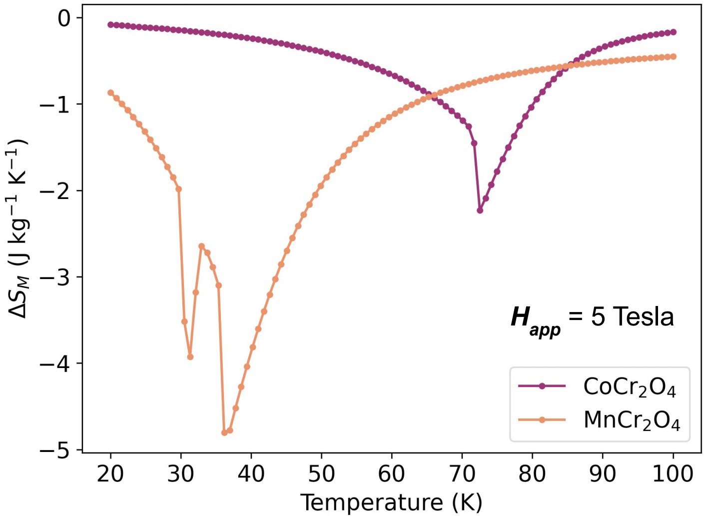
$MnCr_2O_4$

Material test case: $Mn_xCo_{1-x}Cr_2O_4$

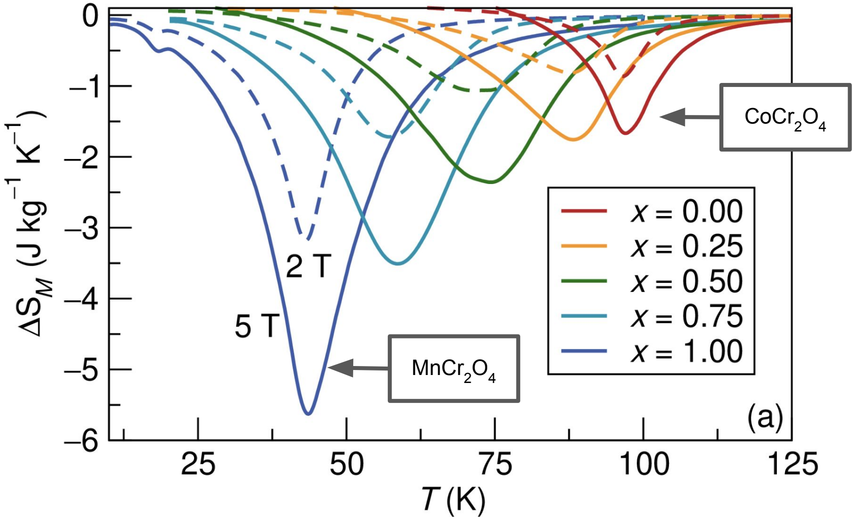
ΔS_M - a magnetocaloric figure of merit

We can successfully predict experimentally resolved thermodynamic behavior

Thermodynamic mean-field model



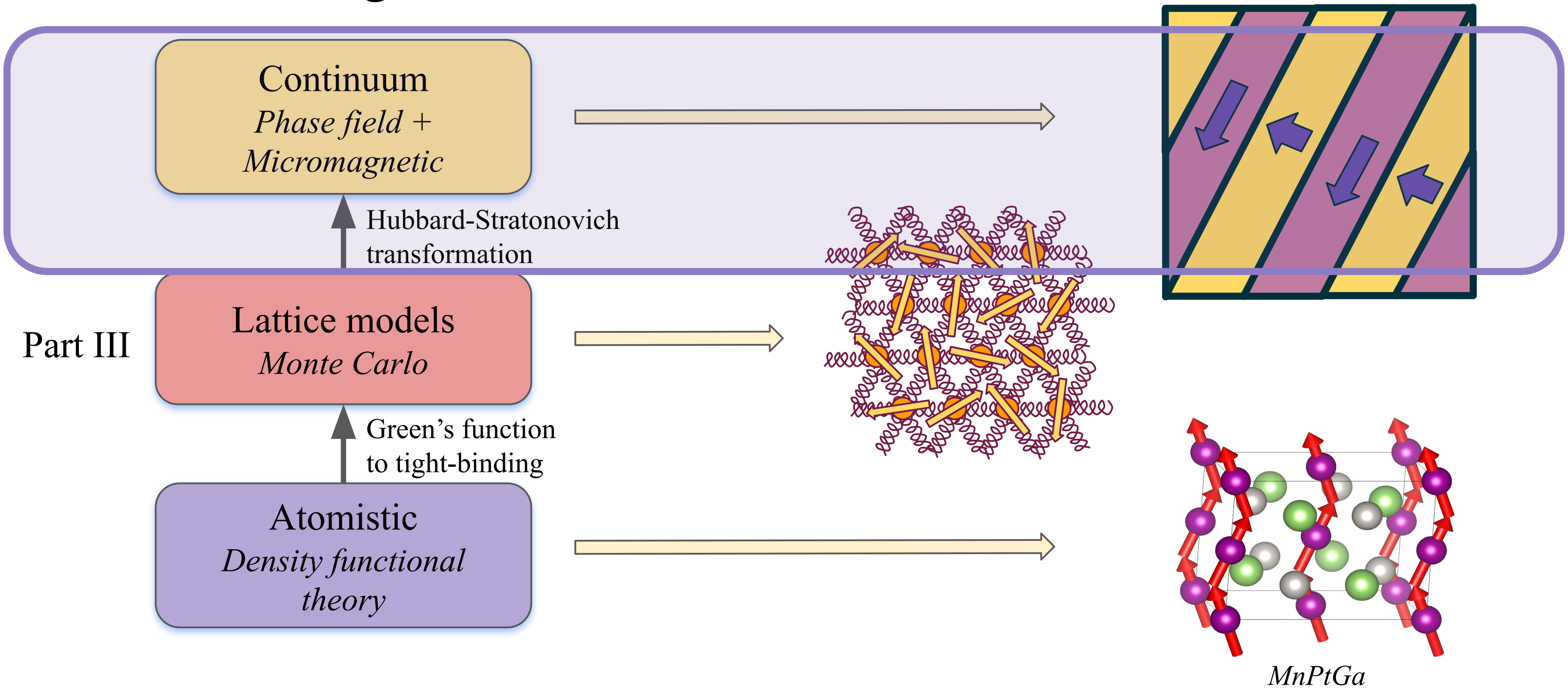
Experimental results
(Joya Cooley et al.)



Material test case: $Mn_xCo_{1-x}Cr_2O_4$

Part IV:
Continuum description

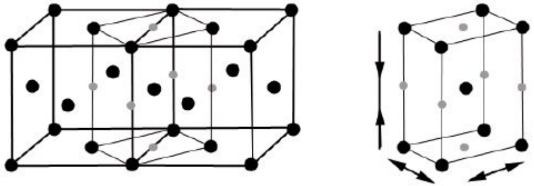
Multiscale magnetic framework



DOE CSGF Practicum at LLNL

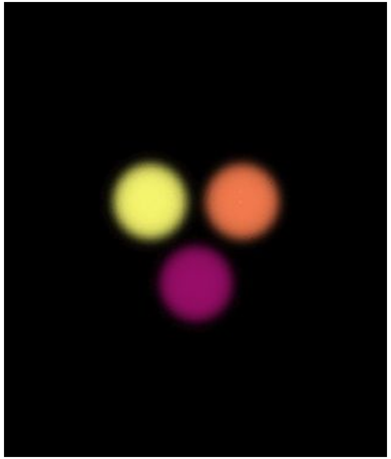
Probing the kinetics and microstructure of diffusionless structural phase transitions using a custom Cython code

- Under the guidance of Dr. Tae Wook Heo
- Fully MPI parallelized

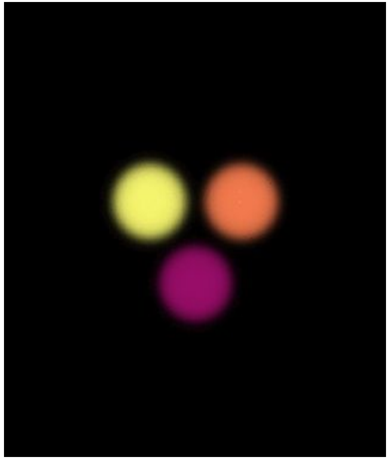


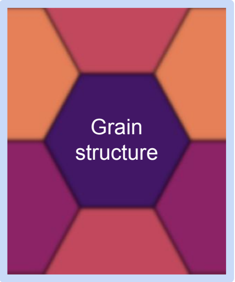
face-centered-cubic (FCC) to body-centered-cubic (BCC)

FCC to BCC



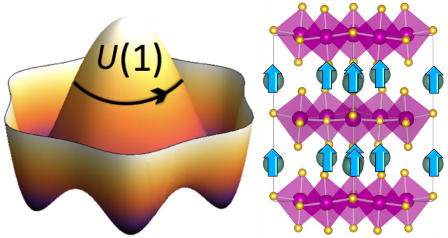
HCP to FCC





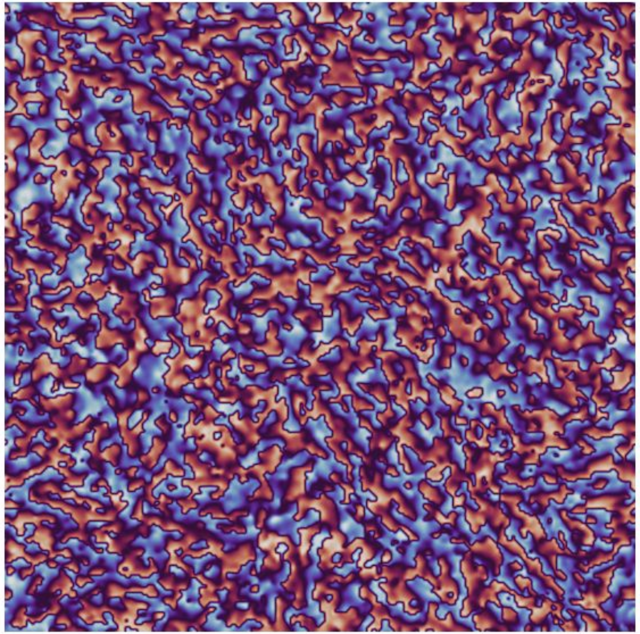
Each color represents a symmetrically distinct structural variant (deformation direction)

Phase field model with random grain structures

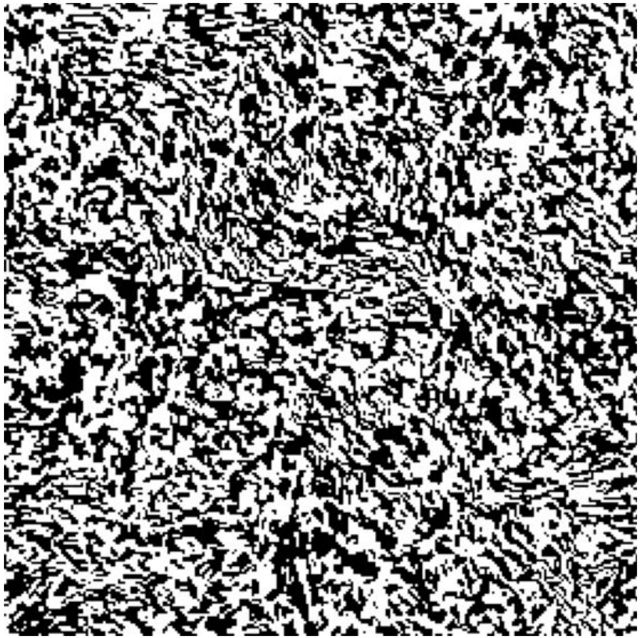


Sinéad Griffin et al.
DOI:10.1103/PhysRevX.2.041022

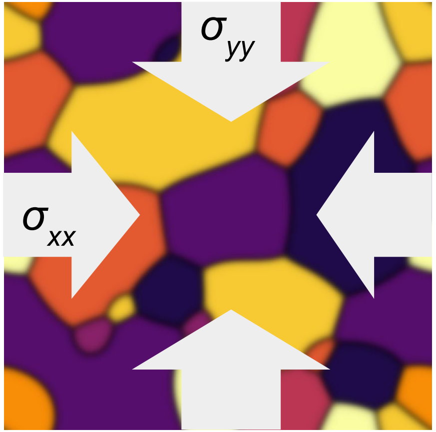
Trimerization OP



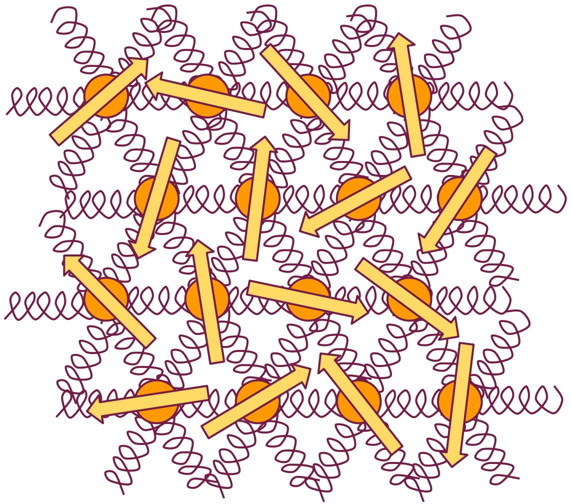
Polarization



Grain structure



Moving to the continuum picture

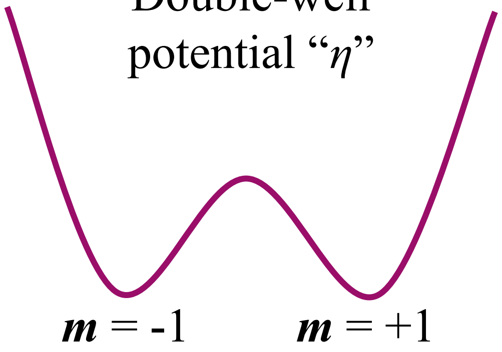


Lattice model

Gaussian integral
"Hubbard Stratonovich"
transformation

Ginzburg Landau functional

Double-well potential "η"



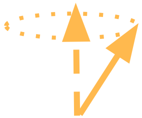
Gradient contributions: $|\nabla \mathbf{m}|^2$

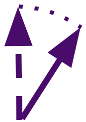
Using a new time evolution equation for micromagnetics

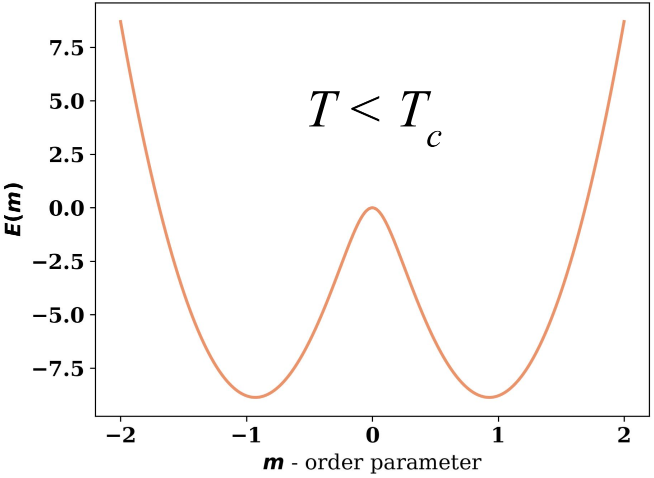
Time evolution is not restricted to $|\mathbf{s}| = 1$

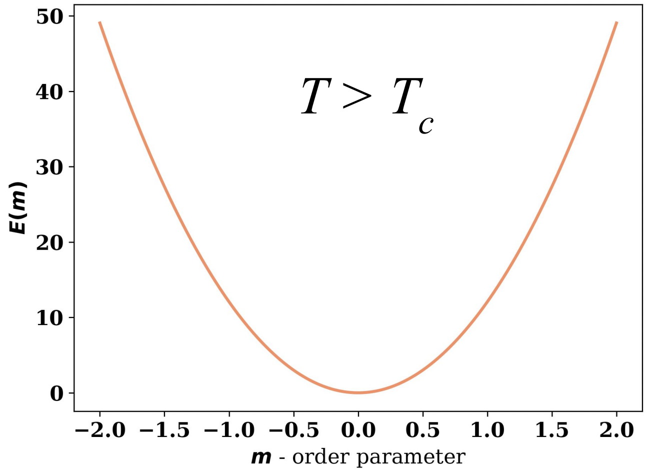
$$\mathbf{h}_{\text{eff}} = -\frac{\delta E[\mathbf{s}]}{\delta \mathbf{s}}$$

$$\frac{d\mathbf{s}}{dt} = -\gamma' \, \boxed{\mathbf{s} \times \mathbf{h}_{\text{eff}}} + L \, \boxed{\mathbf{h}_{\text{eff}}}$$







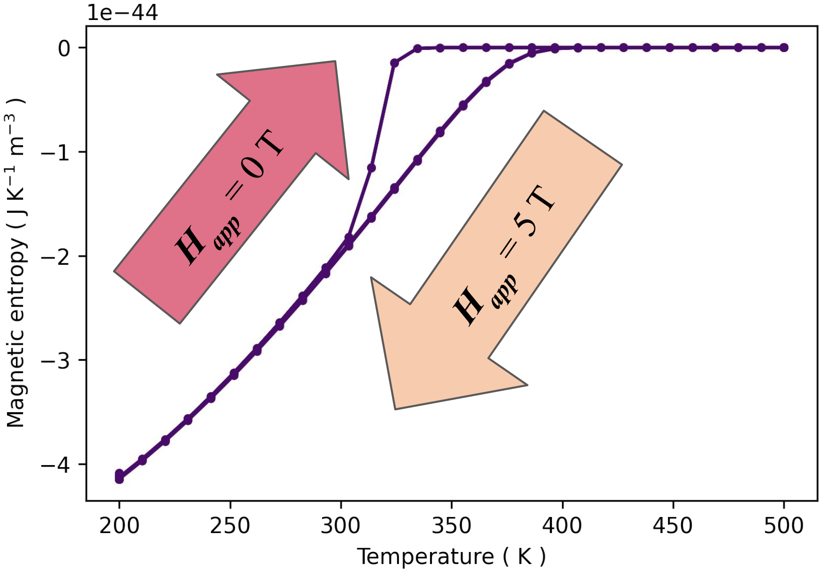


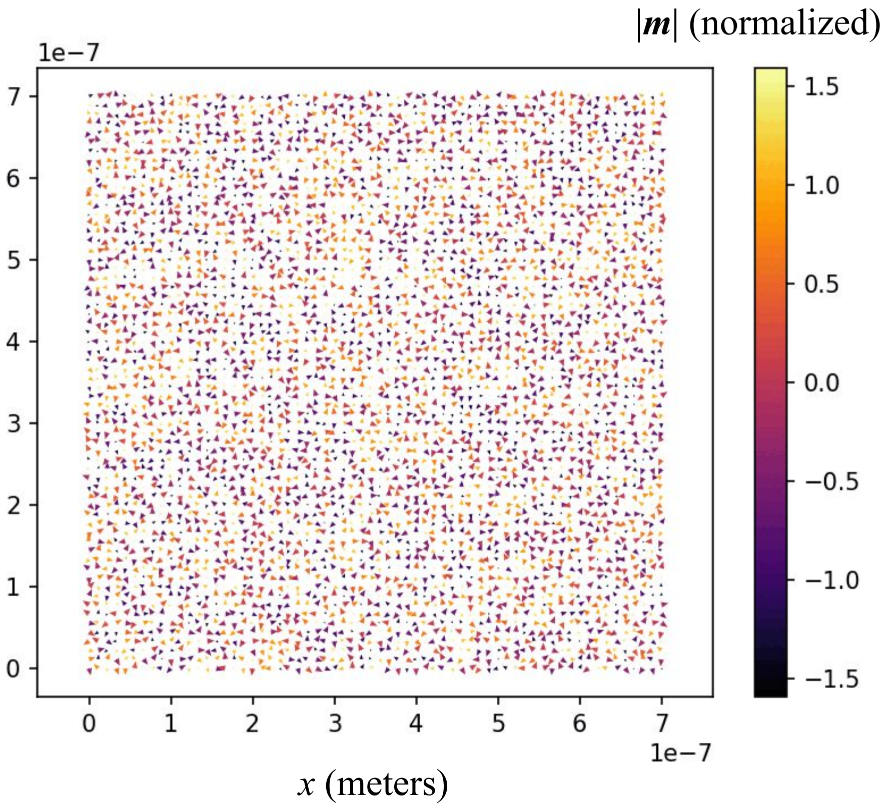
References:

- J. X. Zhang and L. Q. Chen, Acta Materialia, 2005.
- Additional works by L. Q. Chen et al. (and others)

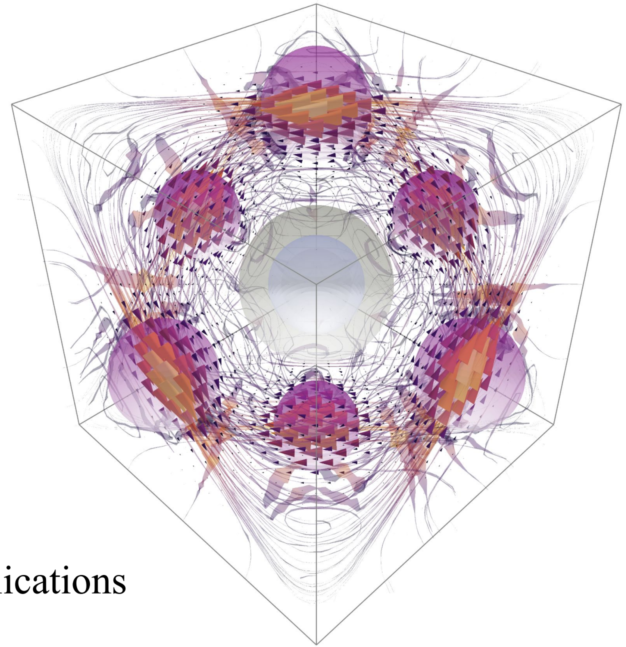
Magnetocaloric cycling

Test case: Ni_2MnGa





Conclusions & Future work



Predicting magnetic properties is challenging

- Magnetic ground-state → many DOFs
- Important: response to applied magnetic field & stress at different temperatures
- Mesoscopic magnetic → full description & contribute to hysteresis - fundamentally non-equilibrium

We address these challenges

- Essential to consider magnetic properties for real-world applications (e.g. magnetocalorics, spintronics, … etc.)
- Combined & holistic approach + many levels of theory *is possible* → Can be successfully applied to predict experimental properties
- Work is transferable to not just explicitly magnetic materials (e.g., predicting U / J → wide range of studies)
- We developed workflows to make this as easy as possible for others to adopt

Thankful of Mentors, Collaborators, and Fellowship

Advisor

Professor Kristin Persson

Mentor

Dr. Matt Horton

Dr. Sinéad Griffin

Dr. Dennis Meier

Isaac Craig

Dr. Jan Schultheiß

Dr. Tae Wook Heo

Professor Joya Cooley

Dr. Edward Linscott

Professor David O'Regan

And others….

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NERSC

BERKELEY LAB