Exploring Pressure and Temperature Induced Magnetostructural Phase Transformations in $Ni_{50}Mn_{50-x}In_x$ Alloys

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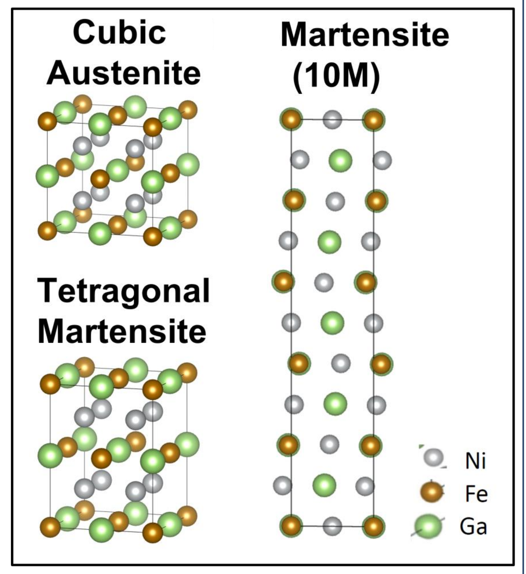
Unraveling Mechanism Mysteries

Crystal structure and martensitic phase transformation

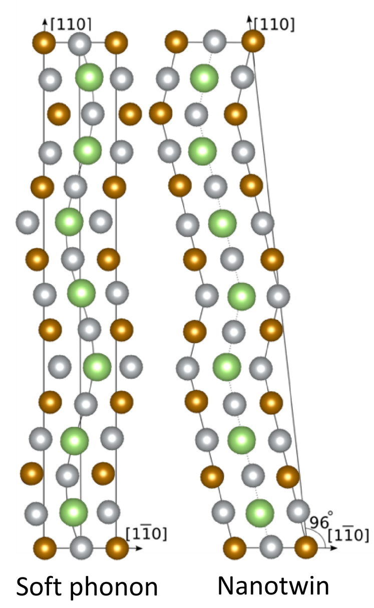
For x≤16, $Ni_{50}Mn_{50-x}In_x$ undergoes a martensitic phase transformation between a high temperature austenite phase (ferromagnetic cubic) and a low temperature martensite phase. For the martensite phase, the magnetic configuration and the presence or absence of modulation are subject to the Indium concentration. The ferromagnetic austenite phase is



What is the mechanism behind the formation of the modulated phase?

Two transformation mechanisms and pathways are considered for forming modulation:

1. Electronic instability arising from soft phonons (physics picture) with continuous sinusoidal modulation
2. Adaptive martensite or nanotwin formation to reduce phase boundary mismatch (mechanics picture) with nanotwin formation

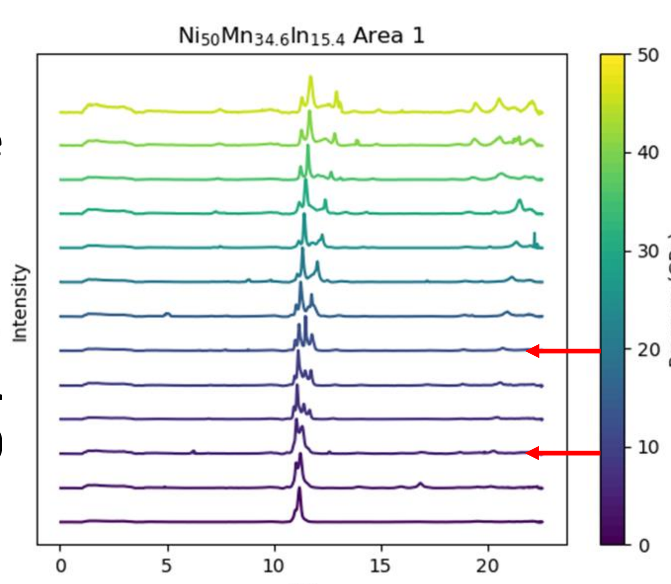


Looking for clues at high pressure

<u>Powder diffraction data for x=15.4</u>

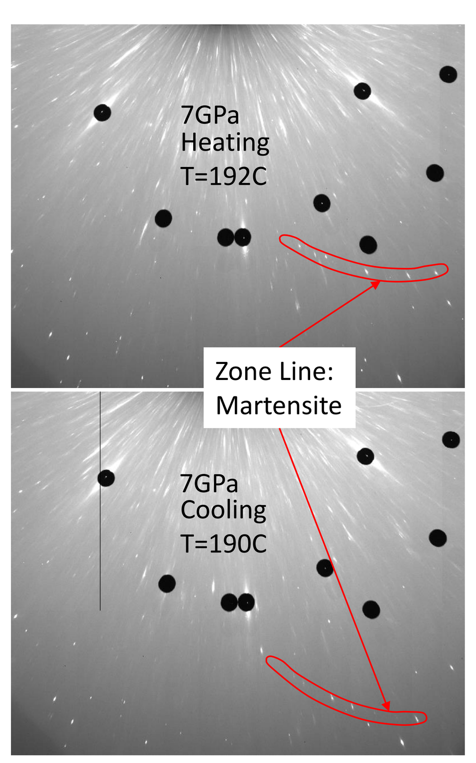
How is the structural transition affecte by pressure?

- Changes in peaks at 7 GPa and 20 GPa indicate phase transformations
- Austenite to martensite near 7 GPa.
- Additional transformation above 20 GPa

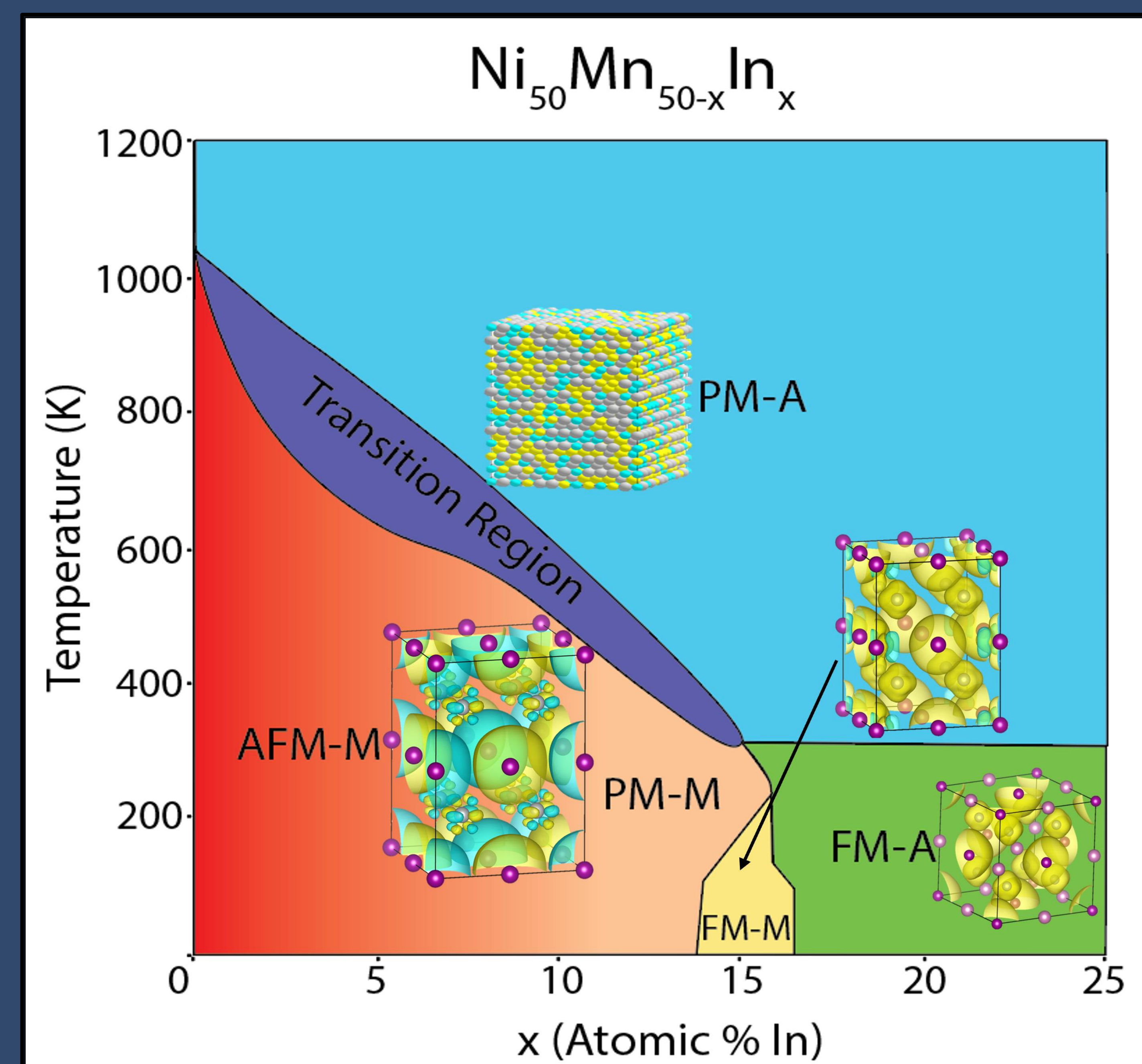


<u>In-situ Laue diffraction with increasing temperature</u>

- Study of modulation formation during isobaric and isothermal phase transformations at 16-BMB at the APS
- White beam Laue microdiffraction just concluded on Feb 13th, with data analysis to follow shortly.
- Our previous attempts were inconclusive regarding modulation but show pressures over 5 GPa stabilize the martensitic phase and raise the transformation temperature
- Done in collaboration with Daniel Shoemaker, Ravhi Kumar, and Rus Hemley



We combine high pressure XRD and first-principles-based models to understand the mechanisms behind magnetostructural phase transformations in $Ni_{50}Mn_{50-x}In_x$



$Ni_{50}Mn_{50-x}In_x$ Is an intriguing material with a rich magnetostructural phase space

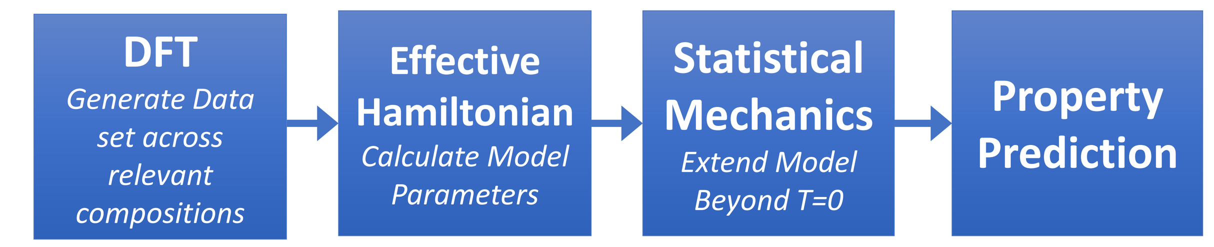
Why study $Ni_{50}Mn_{50-x}In_x$?

- Metamagnetic Heusler Alloy
- Reentrant Ferromagnetism
- Spin Glass
- Shell Ferromagnet
- Shape Memory Alloy
- Magnetocaloric Effect
- Electrocaloric Effect
- Barocaloric Effect

Modeling Magnetic Behavior

The magnetic structure is an important component of the overall phase stability and many potential applications for $Ni_{50}Mn_{50-x}In_x$. We have developed a combined cluster expansion and Ising-like model to study the magnetic characteristics in a given local chemical environment. The model is parametrized solely from first principles calculations.

<u>Computational Workflow</u>

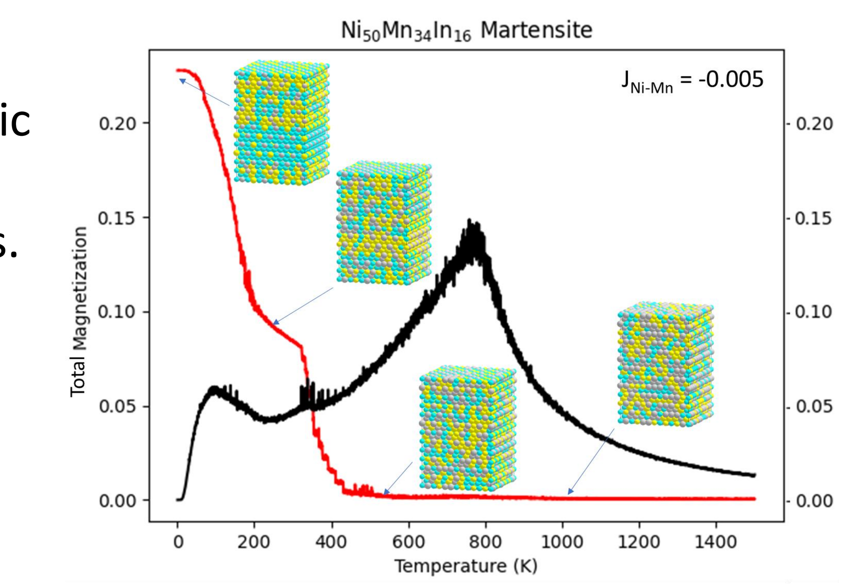


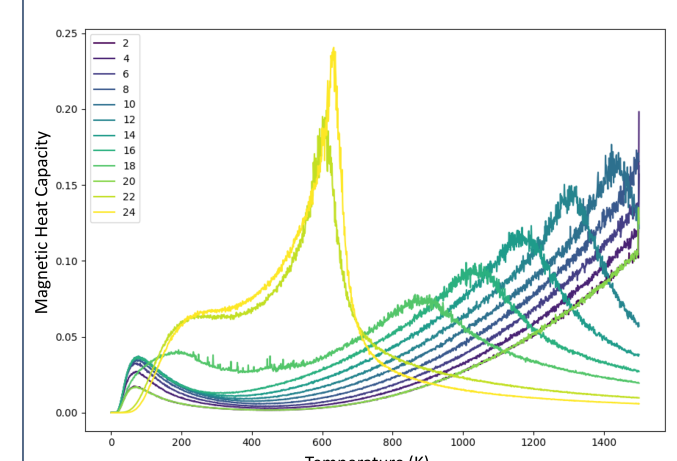
<u>Effective Hamiltonian</u>

$$H = \sum_{\alpha}^{A\in(n'+n)} J_{\alpha}^{c}\Phi_{\alpha}(\sigma) + \sum_{i,j}^{n} J_{i,j}^{s} s_i s_j + \mu \sum_{i}^{N} h s_i + NC$$

Thermodynamics and phase transformation behavior

- Our effective Hamiltonian offers a rich set of magnetic transitions consistent with experimental observations.
- Ni-Mn nearest neighbors play an important role in mediating competing FM and AFM exchange interactions



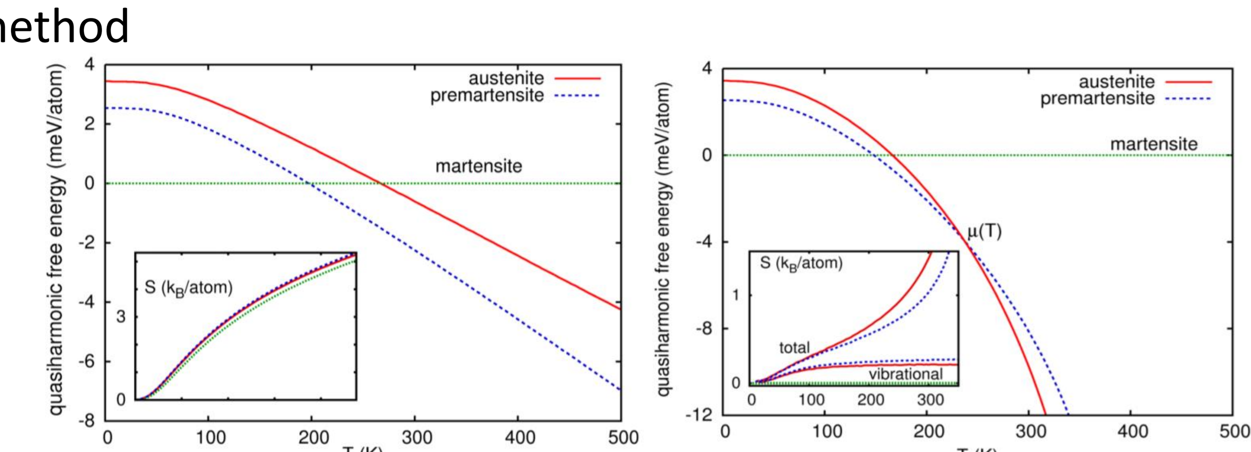


- Tuning the alloy stoichiometry shows an abrupt change in the magnetic transformation behavior above x=18.
- This highlights the sensitivity of the alloy system to stoichiometry and short-range order/disorder

Combining magnetic and vibrational contributions

We are exploring methods for integrating magnetic-configurational free energy with vibrational contributions. These include:

- Alloy generalization with the coherent potential approximation
- Supercell phonon unfolding
- Accounting for phonon magnon interactions using the fixed moment method



Uijttewaal, M. A., et al. "Understanding the phase transitions of the Ni2MnGa magnetic shape memory system from first principles." *Physical review letters* 102.3 (2009): 035702.