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Pressure-dependent thermodynamics of cubic Lu-H-N solid solutions by Monte Carlo simulations based on graph neural networks

Pinwen Guan, Matthew D. Witman, Catalin D. Spataru, Vitalie Stavila, Peter A. Sharma
Sandia National Laboratories

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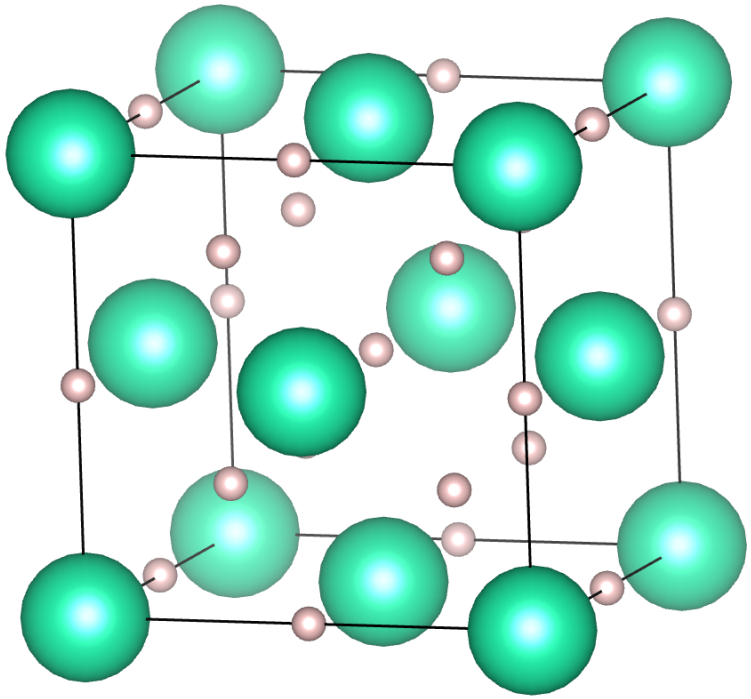
Cubic Lu(H,N,Va) phase

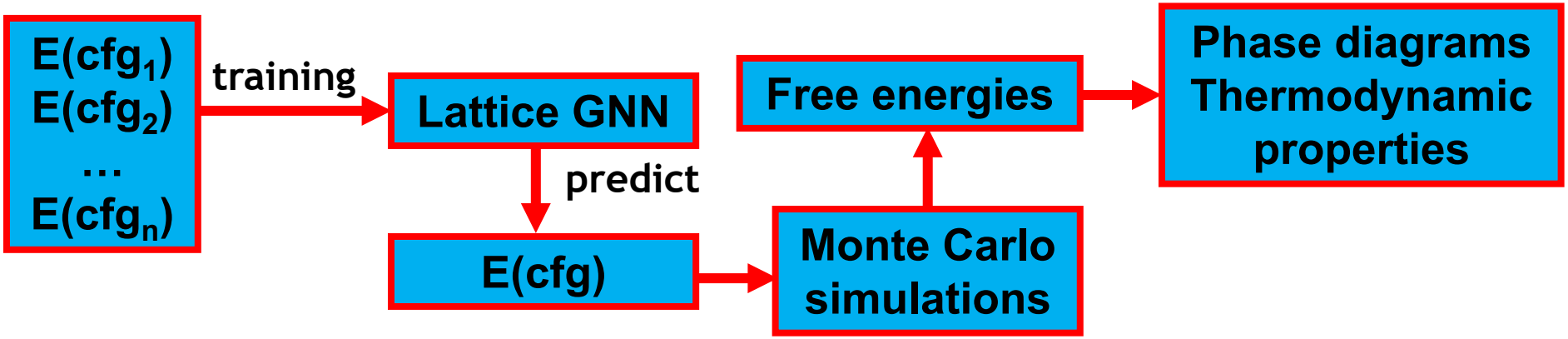
Green: Lu Pink: H,N,Va



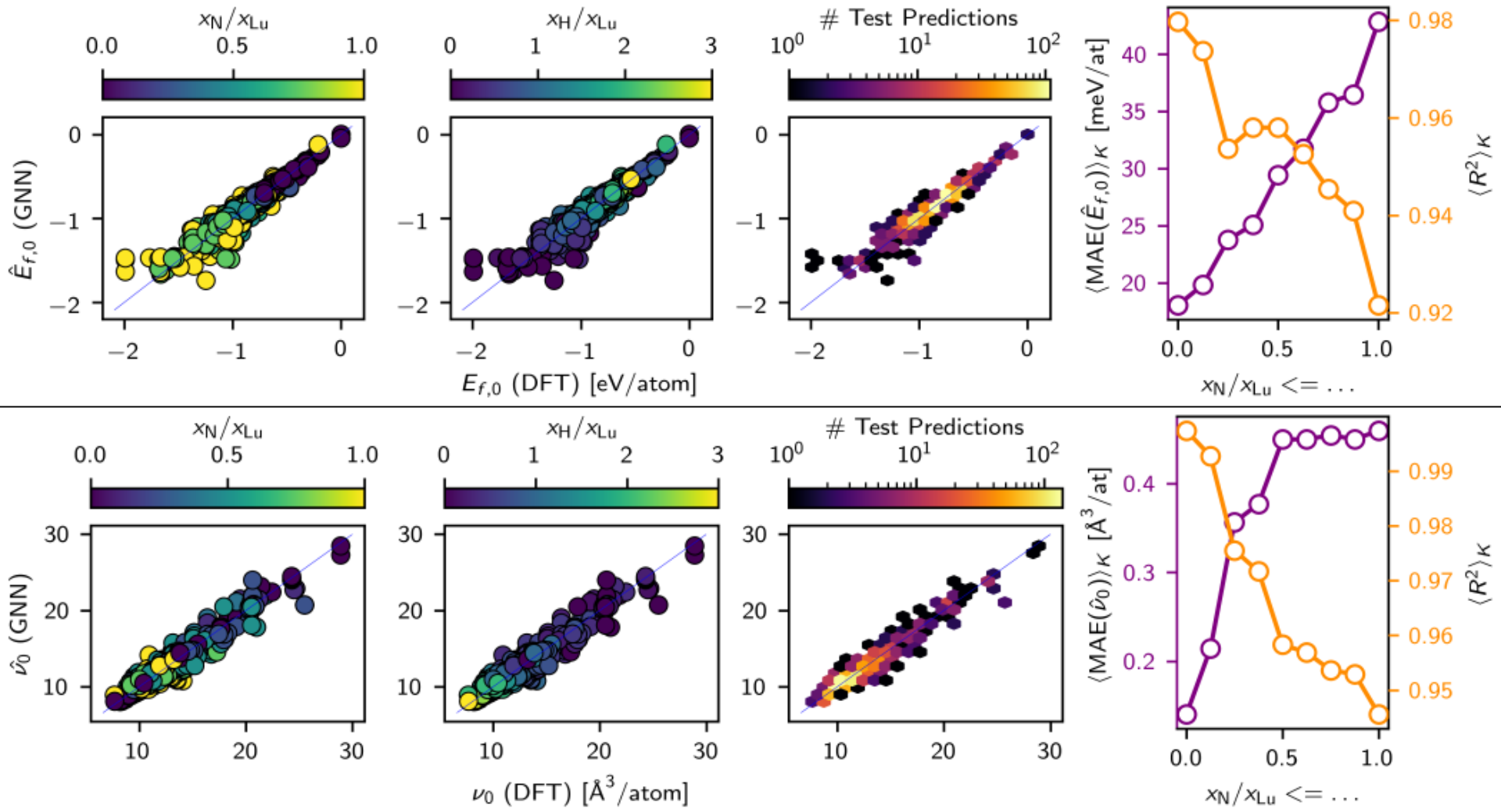
Lu atoms form an fcc lattice, with interstitial sites occupied by H, N or vacancy, forming numerous configurations

Previously, thermodynamics of a solid solution was usually studied using cluster expansion (CE) method, which is challenging for high compositional complexity systems.

The primary goal here is to explore the GNN+MC approach as an alternative of CE+MC.



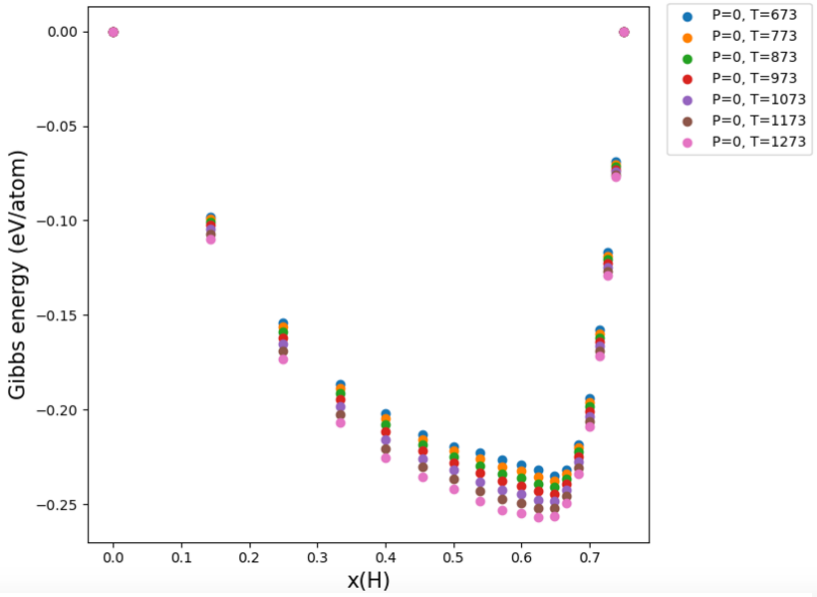
GNN models of configurational properties

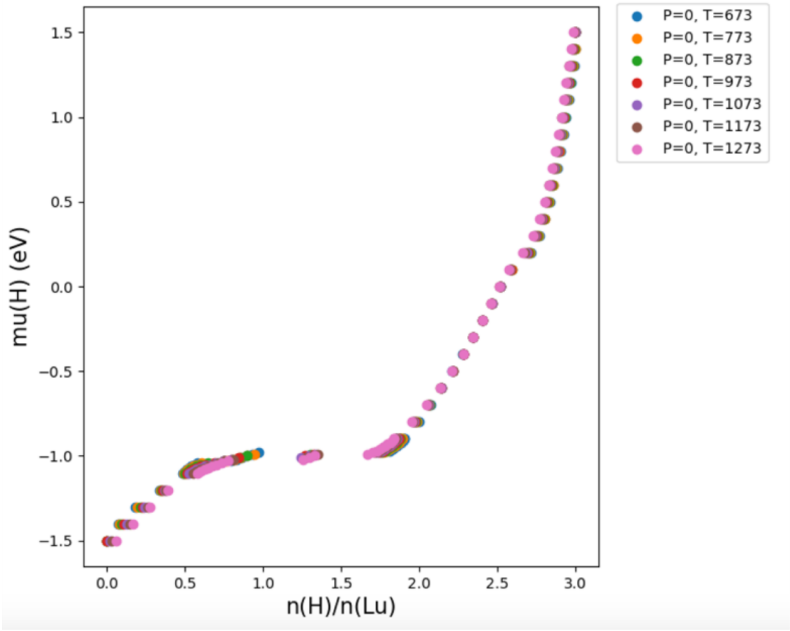


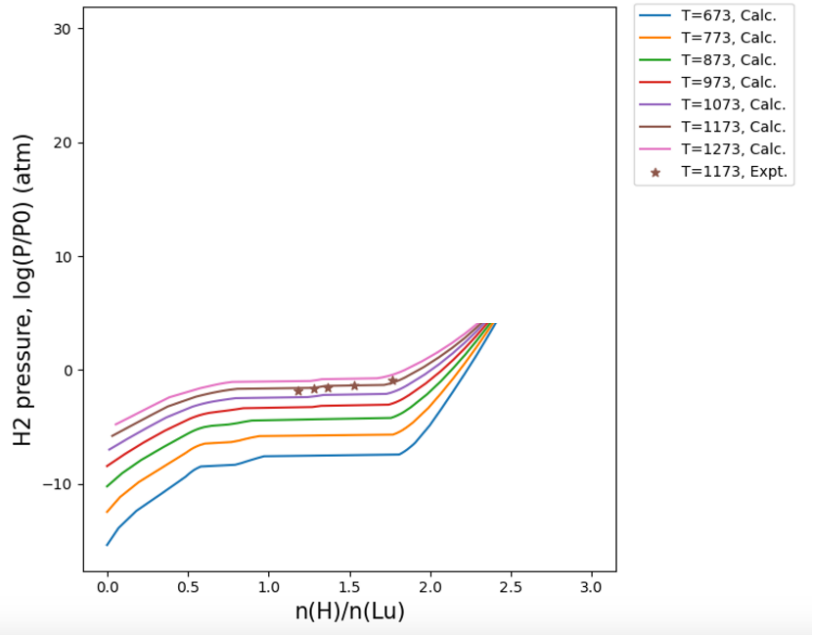
The DFT-calculated enthalpies can be well described by $H=E_0+PV_0$ up to several GPa. Thus, GNN models are built for (E_0,V_0).

The model achieves chemical accuracy (4 kJ/mol) when x(N)/x(Lu)<1.

GNN+MC results for Lu-H (near-ambient)

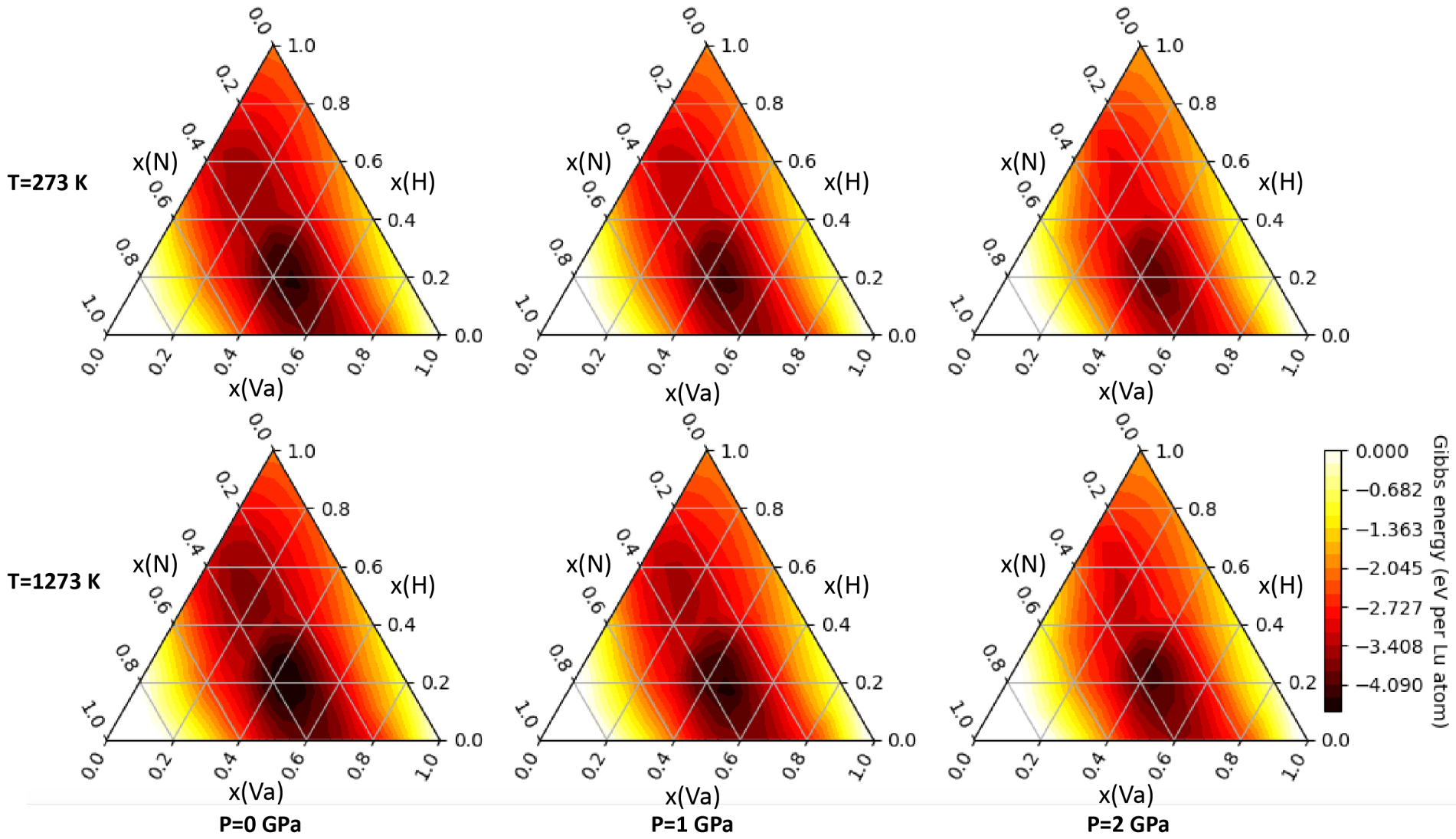






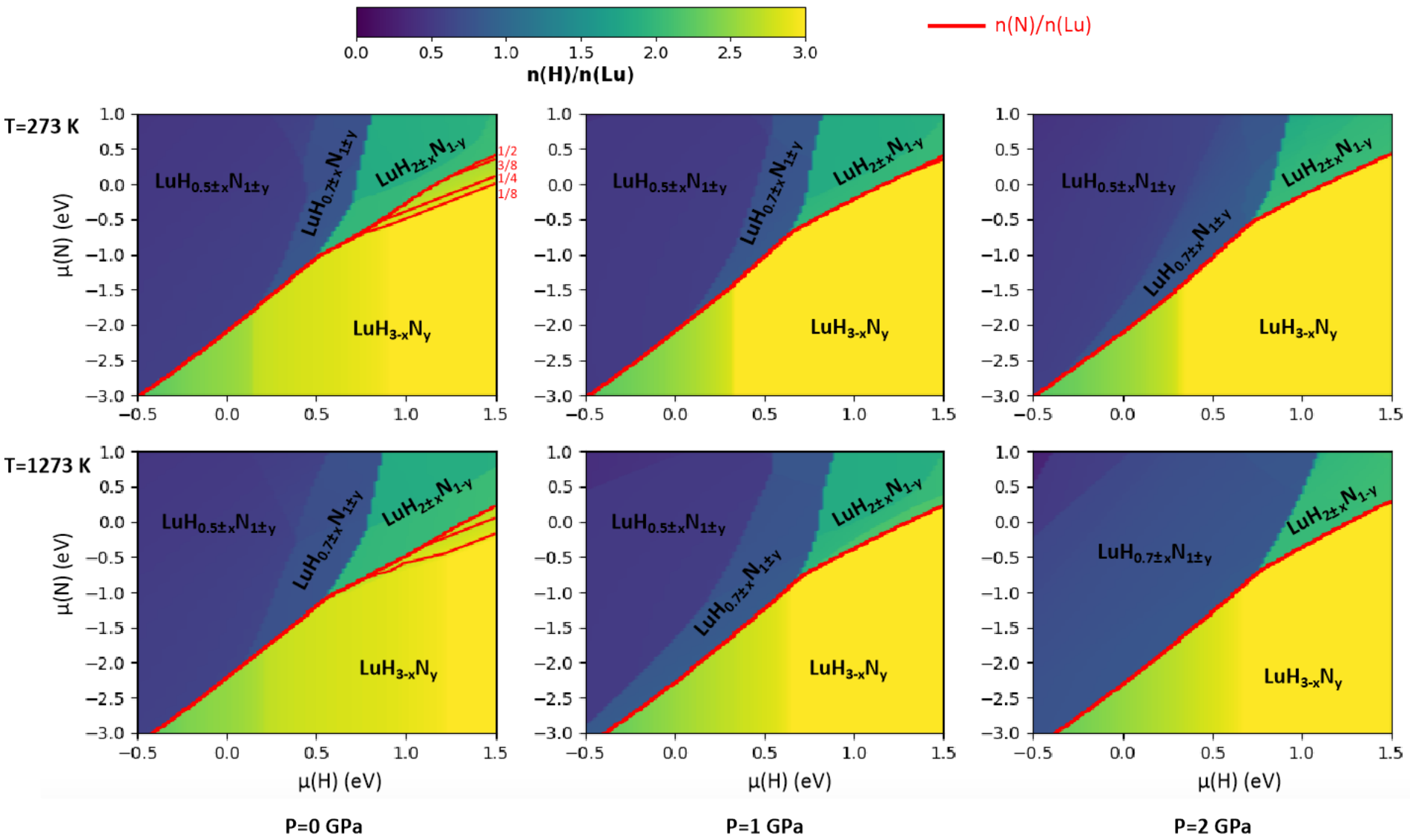
These are the only reliable experimental data to compare with. The calculated PCT curves show good agreement with experiments, demonstrating validity of the methodology.

Cubic Lu-H-N phase Gibbs energies



No vibrational contributions have been considered. When pressure increases, the H-rich region changes most due to high volume per atom.

Cubic Lu-H-N phase diagrams



It's difficult to achieve significant N-doping levels in $LuH_{3-x}N_y$ under equilibrium, especially when pressure increases.

Thanks for your attention!