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Title: Phonons as a means for understanding Pu-Ga materials

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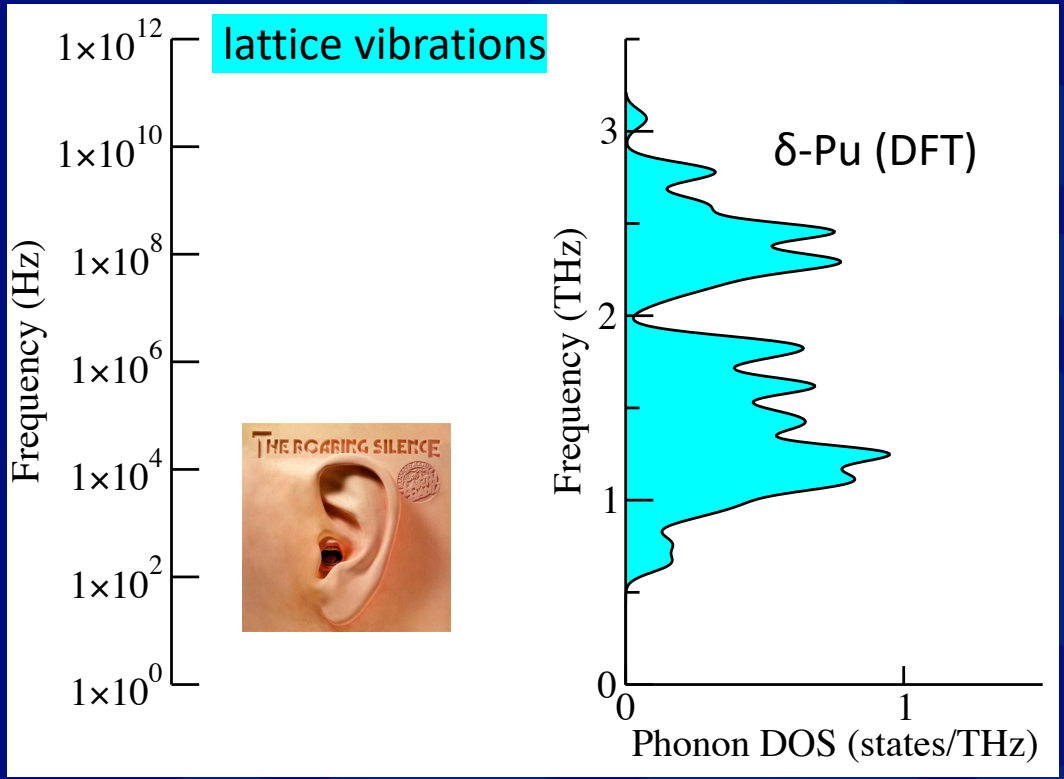
Los Alamos NATIONAL LABORATORY

Phonons as a means for understanding Pu-Ga materials

Sven P. Rudin, T-1

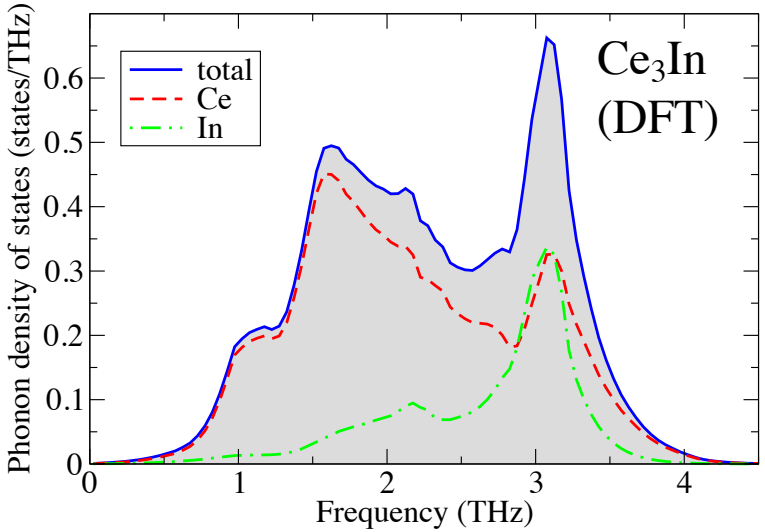
December 14, 2021

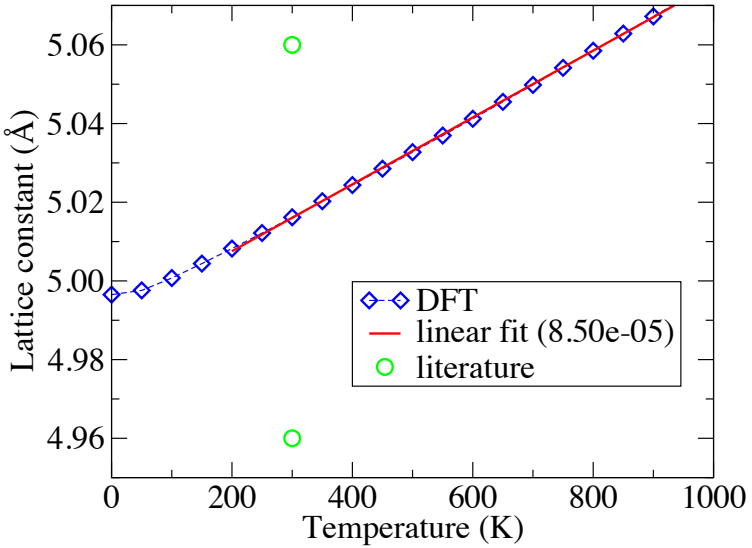


NNSA

LDRD

DFT: Ce_3In as "cold" system to align experiment and theory – change *f* electron treatment?

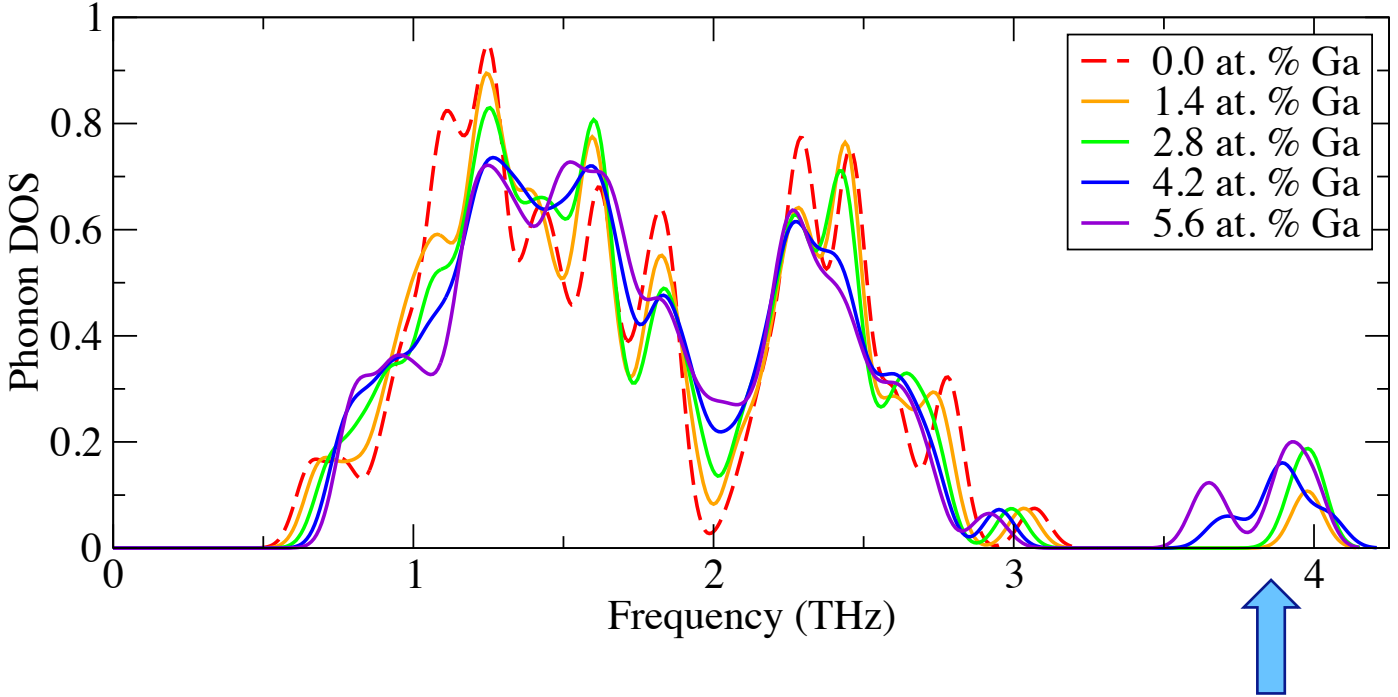




Heat capacity… previous talk:
cannot ignore electronic contribution.

Theory (LANL)	a at 300 K	5.02
	da/dT	8.5E-05
	$1/a\ da/dT$	1.693E-05
Experiment (LANL)	$1/a\ da/dT$	1.245E-05

DFT: Ga doping of δ-Pu introduces phonon modes at experimentally observed frequencies.

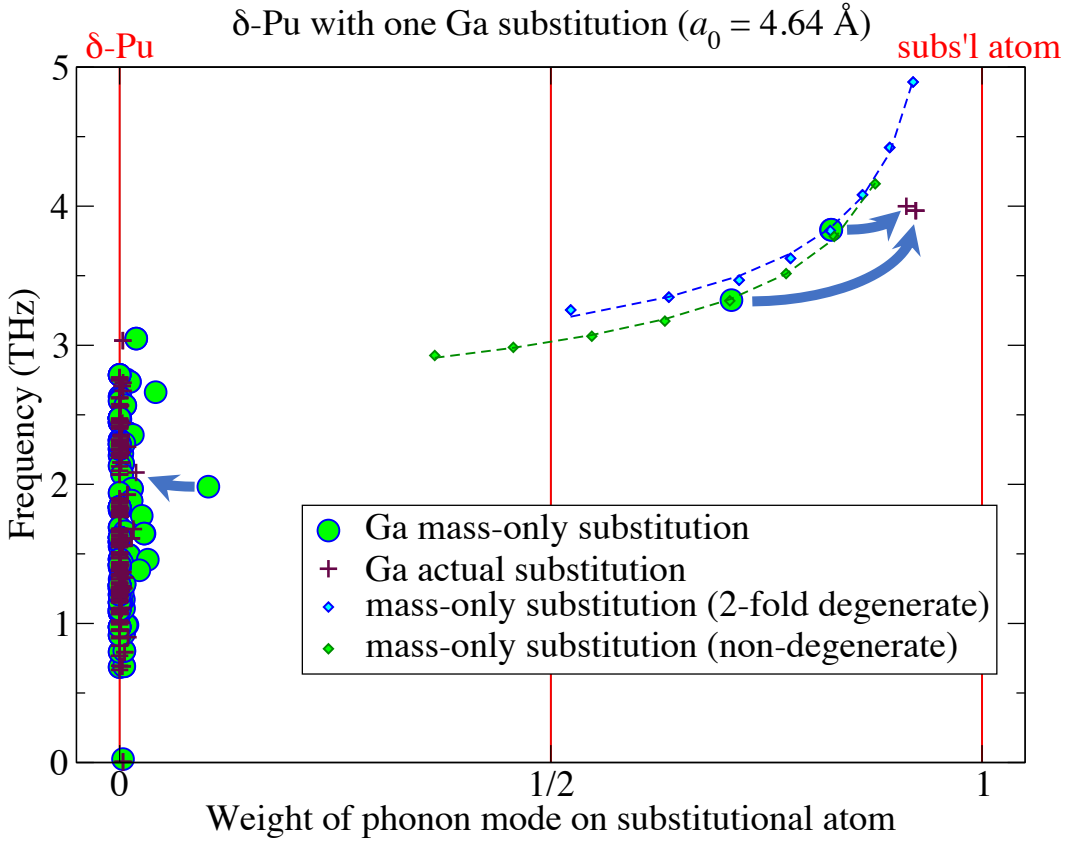


Measured for Ga in Pu-3.6 at. % Ga: $f_{average}$ = 3.96 THz

LYNN et al. PRB 58, 11414 (1998)

new modes (interacting Ga atoms at higher concentrations)

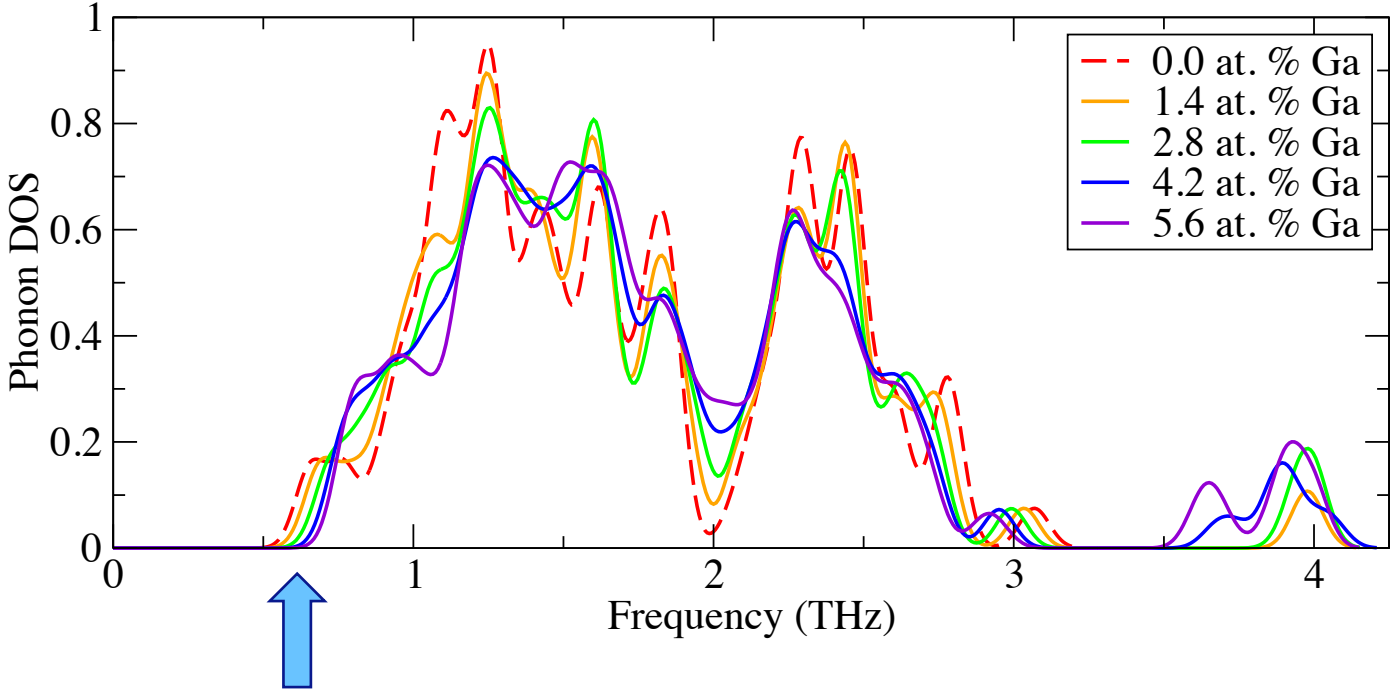
DFT: Ga doping of δ-Pu introduces phonon modes that are Ga-dominated and localized.



Compared to mass-only substitution:

1) Pu-dominated modes
2) Ga-dominated modes
   a) more localized
   b) higher frequencies

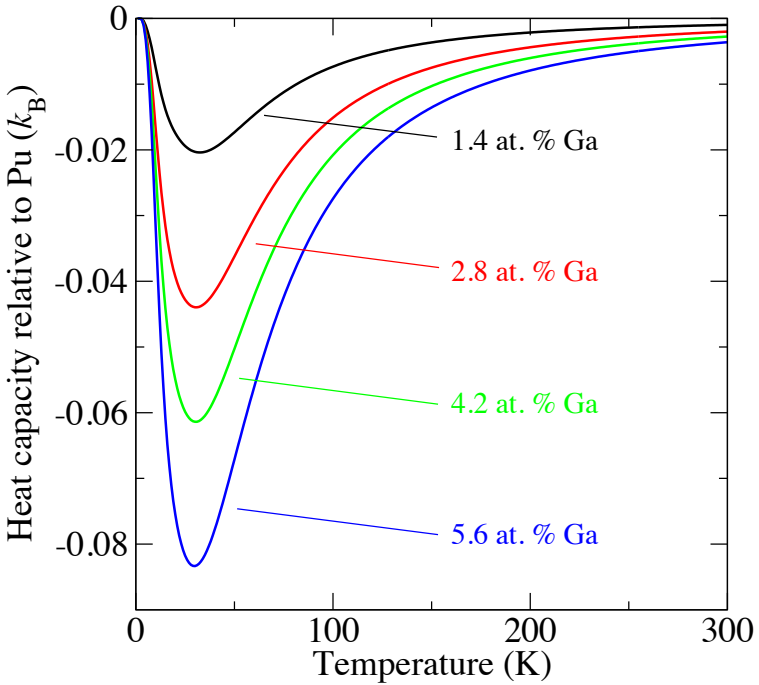
DFT: Ga doping of δ-Pu suggests stabilization of delta phase is not thermodynamic.

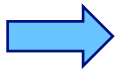


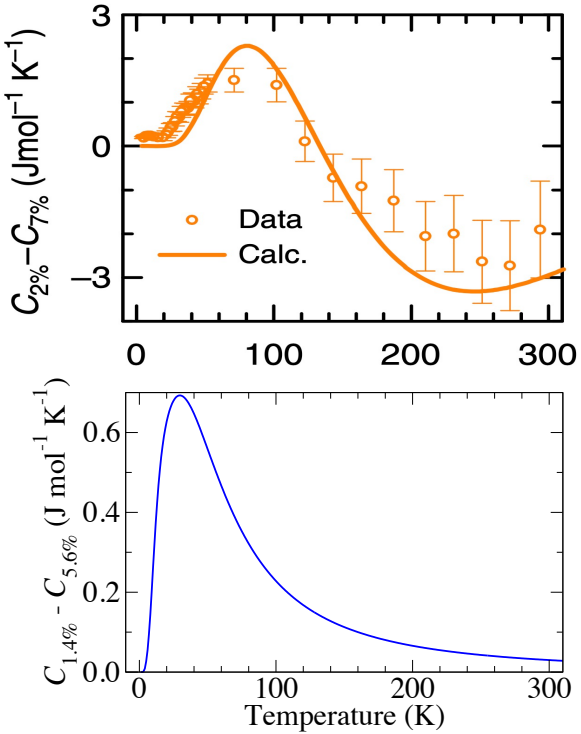
stiffening
of low-frequency modes

- thermodynamics: delta phase less stable
- proposed pathways to alpha: more stable

DFT: Ga doping of δ-Pu changes the phonons' contribution to the heat capacity. (But the effect is swamped by the electronic contribution.)



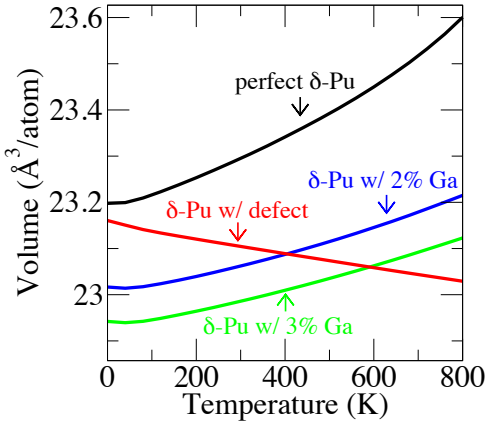




HARRISON et al. Nat. Comm 10, 3159 (2019)

Next steps

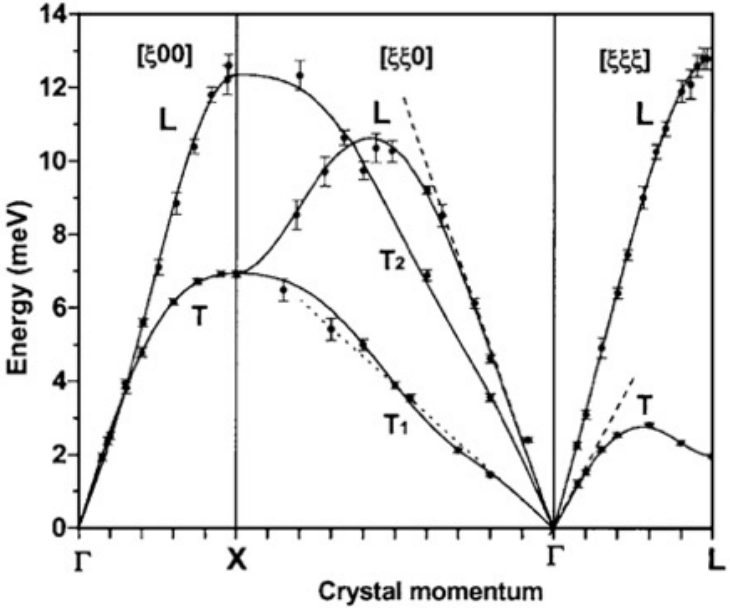
1. Ce_3In: modify treatment of f electron in calculations.
2. DFT calculations of Ga-doped alpha-Pu: how does alpha-Pu's thermodynamic stability change?
3. Repeat the calculations with Al and In instead of Ga: do trends emerge consistent with experimental observations?
4. Explore with DFT the energy landscapes defined by the phonons proposed to initiate α-to-δ transition pathways: what do the landscapes look like? Does Ga doping change them?
5. DFT calculations of the effect of Ga doping and interstitial defect: how does Ga affect the instability induced by the defect?



DFT: effect of magnetic structure on phonon modes

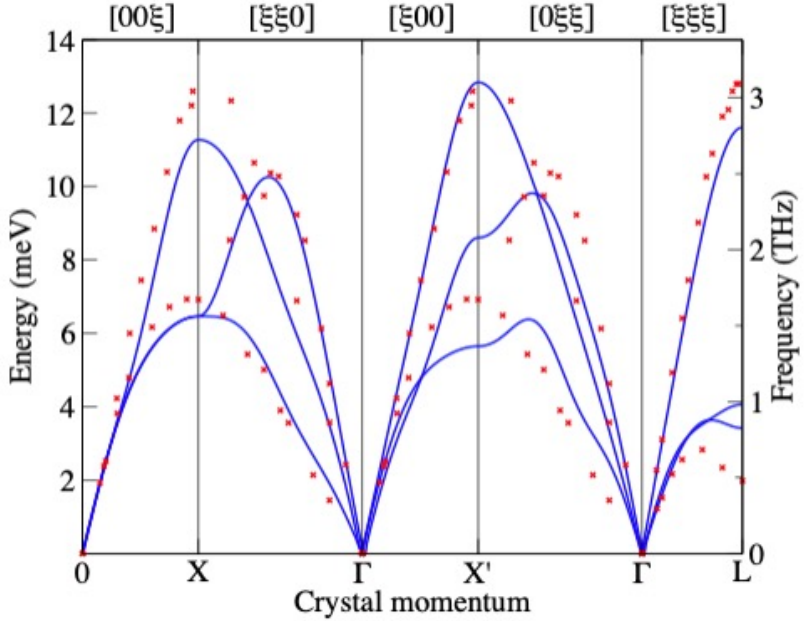
Pu-2 at. % Ga

WONG *et al.* Phys Rev B **72**, 064115 (2005)
Inelastic x-ray scattering

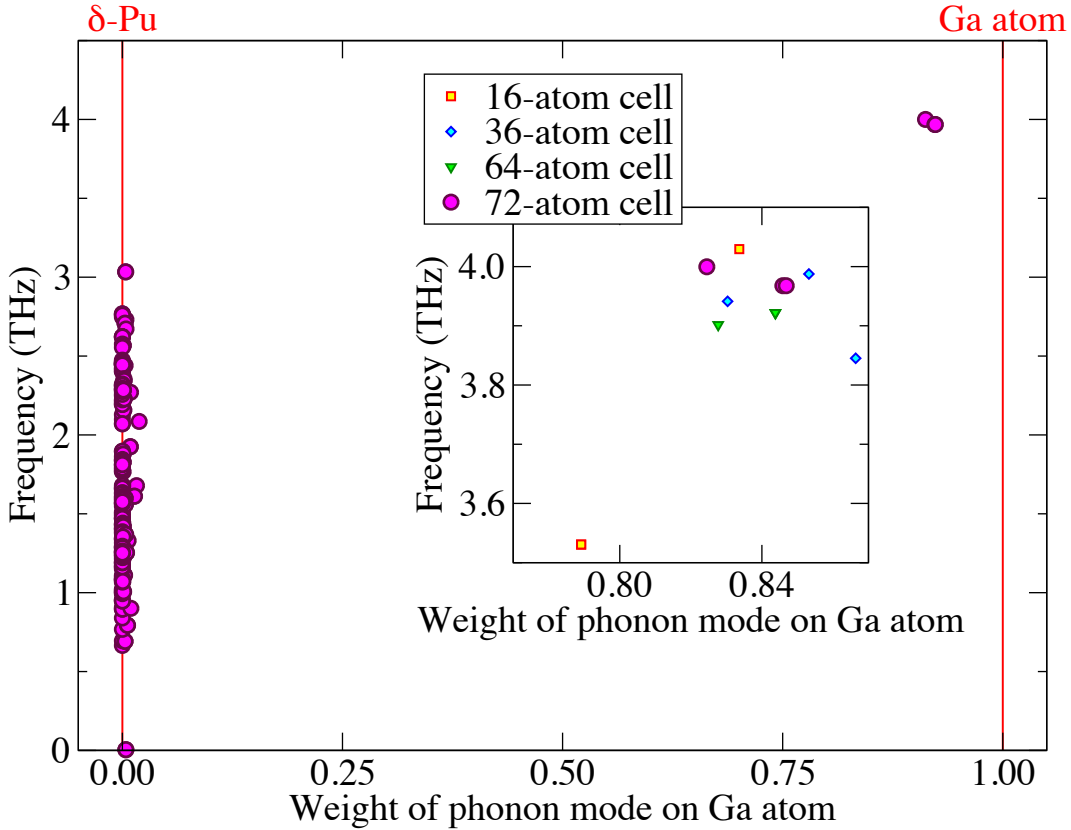


Pu-0 at. % Ga

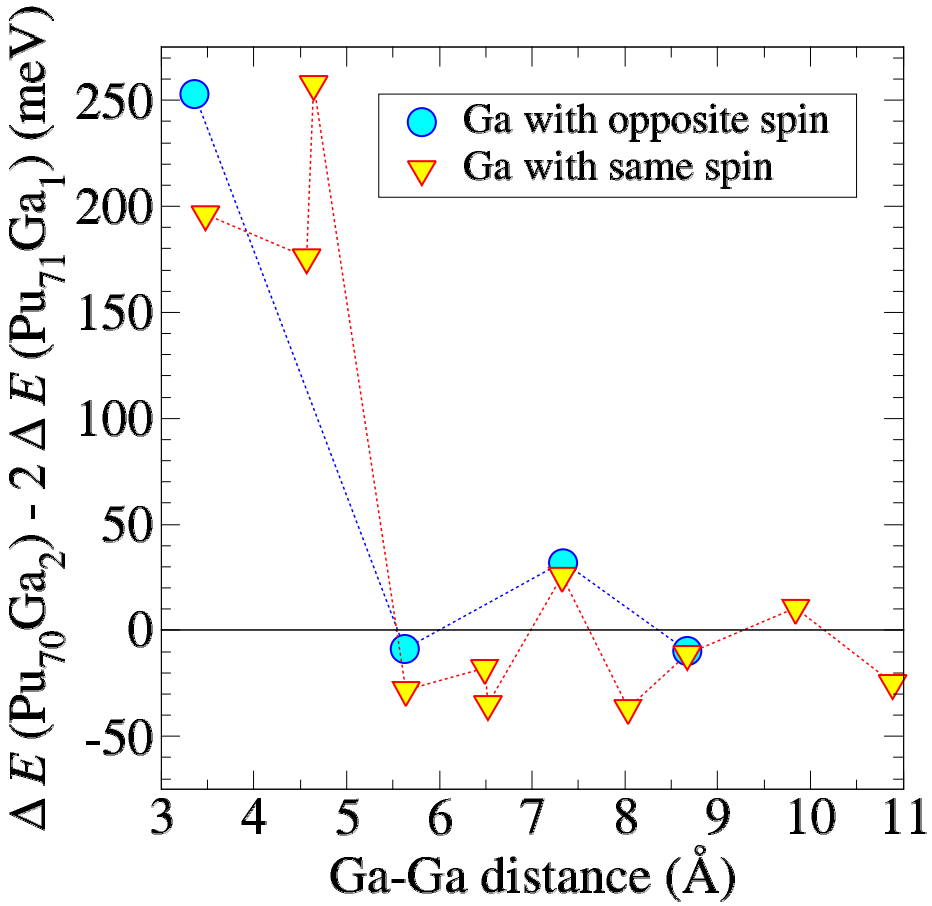
DFT (AFM)



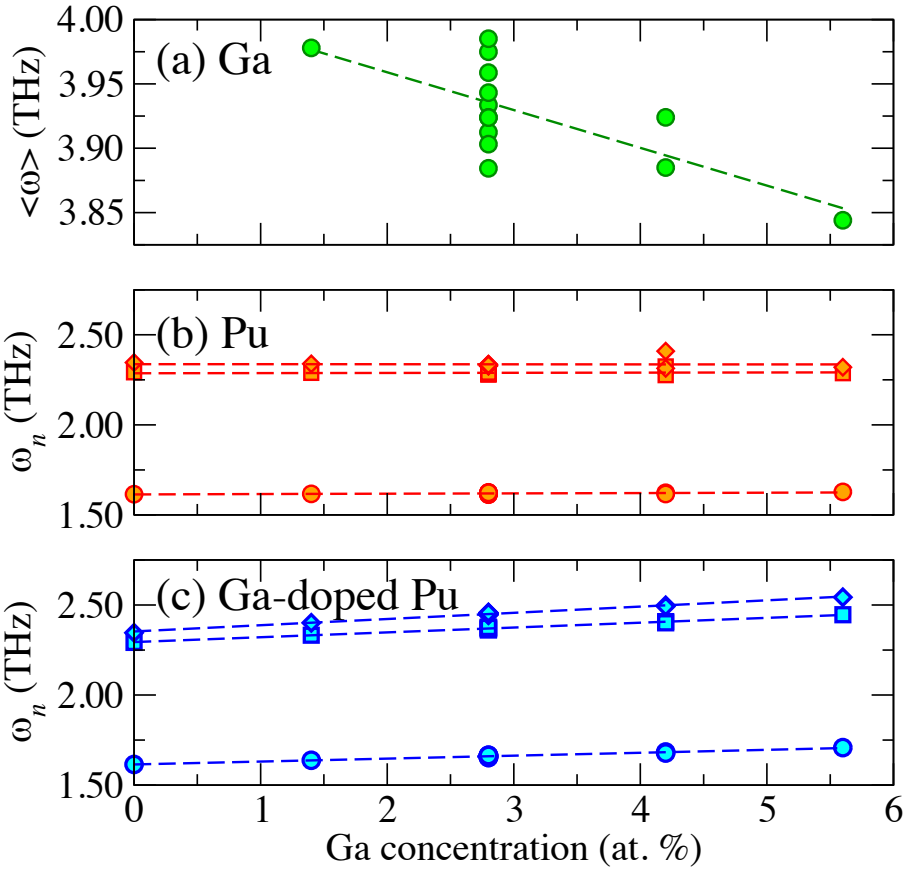
Choice of 72-atom cell size



Range of Ga-Ga cold interaction in 72-atom cell



Phonon moments: change with Ga concentration



DFT: effect of Ga doping on character of low-frequency phonon modes