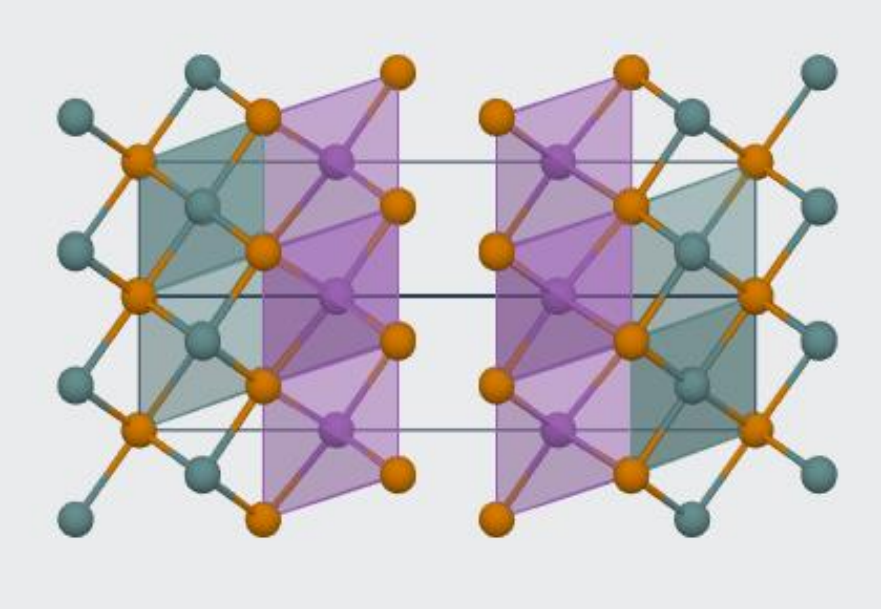
SAND2022-2241C

Atomic Simulations of GST+C Phase Change Materials



Robert Appleton, Zachary McClure,
Alejandro Strachan, David Adams
rapplet@purdue.edu, zmcclure@purdue.edu,
strachan@purdue.edu, dpadams@sandia.gov
School of Materials Engineering
Purdue University
West Lafayette, Indiana USA

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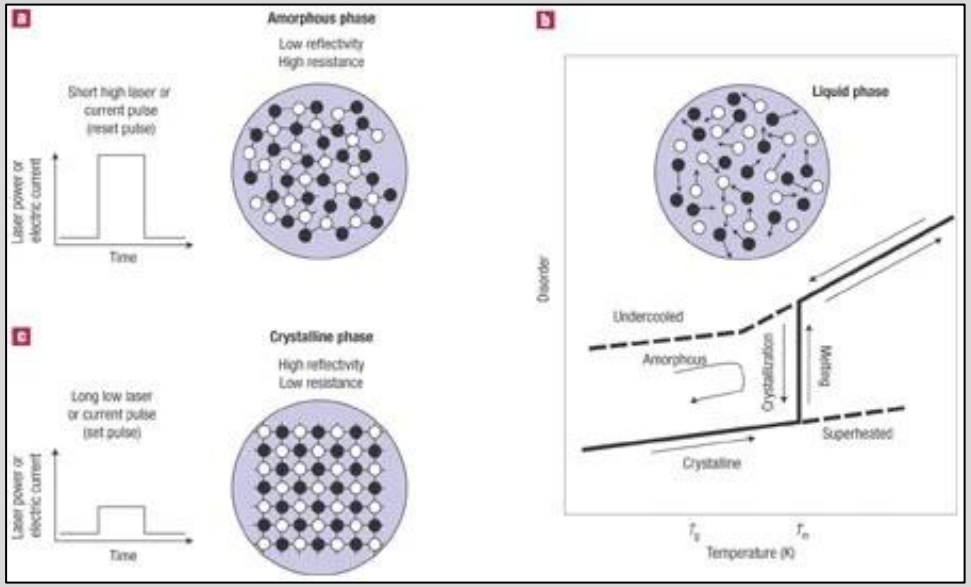
Phase-Change Materials for Data Storage

Store information in physical structure

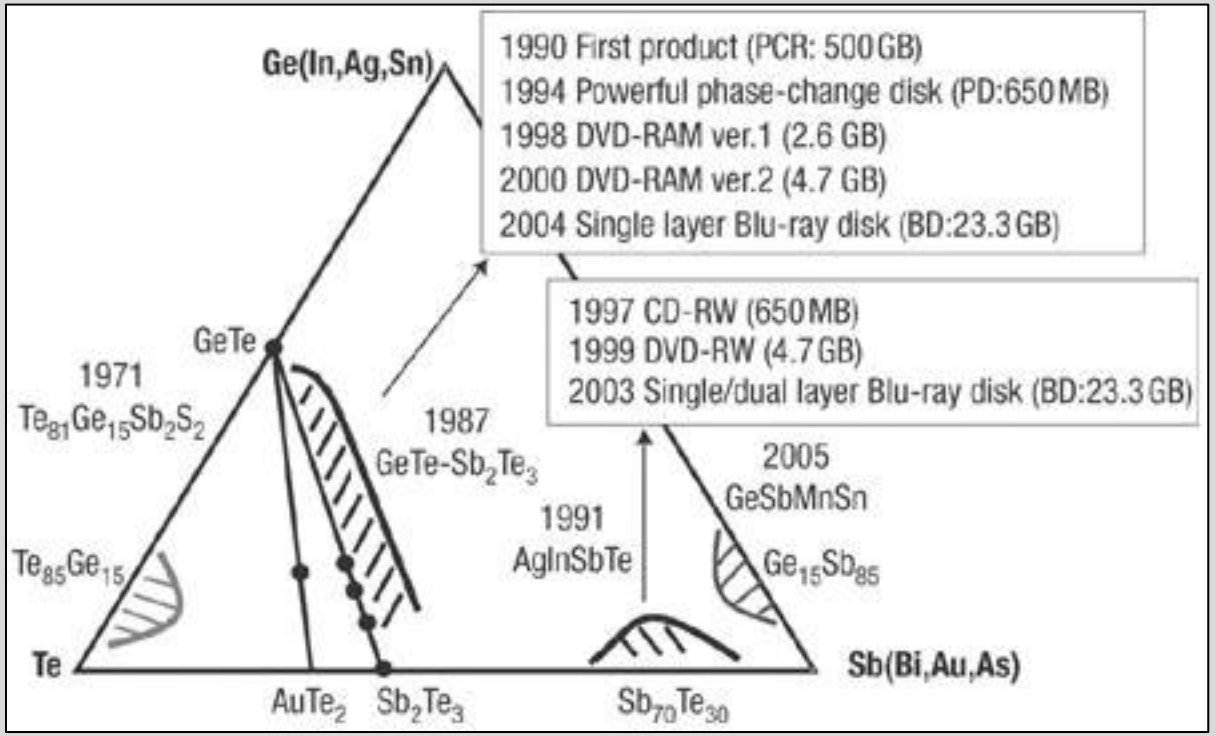


Phase-change materials for rewriteable data storage, M. Wuttig & N. Yamada, *Nature Materials*, 6, 824-832 (2007)

Advantages over flash memory

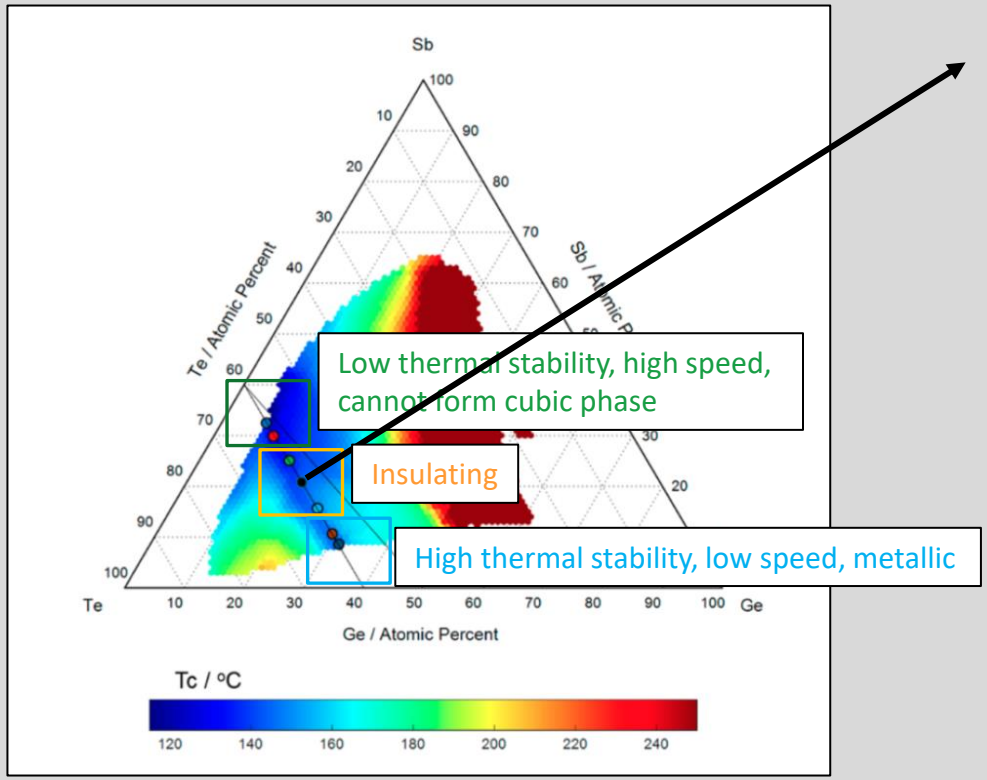
- Increased switching speeds (Flash ~100µs and PCM ~ns)
- Higher storage stability
- Size reduction (nano scale devices)

History of first PCMs and their uses

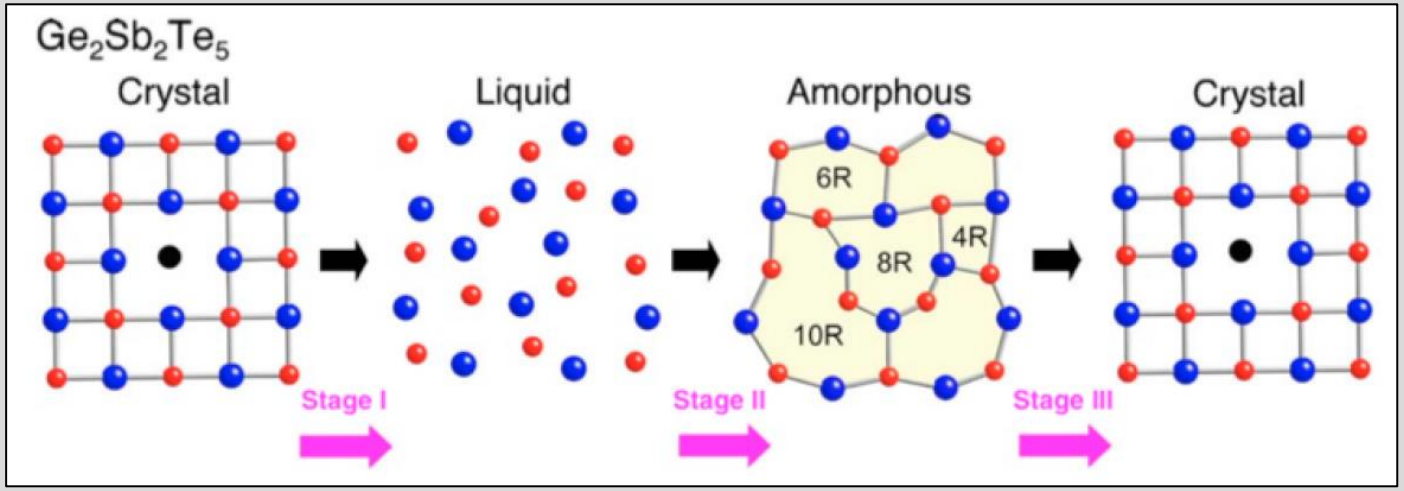


GeSbTe (GST) Alloys

Most prevalent class of PCM due to its fast and reversible phase transitions between amorphous and crystalline states.



Synthesis and screening of phase change chalcogenide thin film materials for data storage. S. Guerin, B. Hayden, D. Hewak, C. Vian, ACS Comb. Sci. 2017, 19, 478–491.

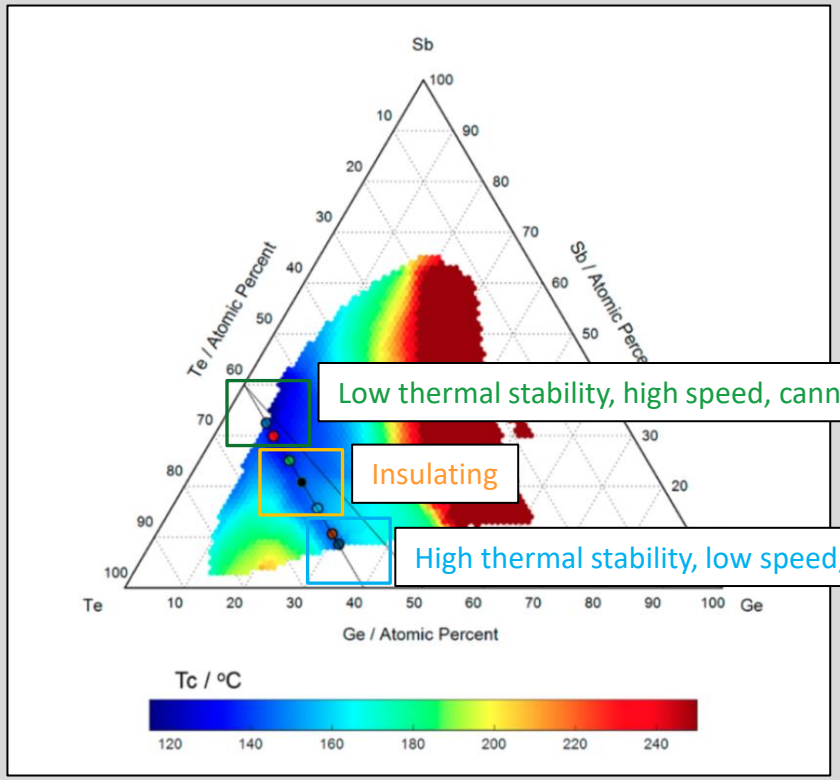


Structural basis for the fast phase change of Ge2Sb2Te5: Ring statistics analogy between the crystal and amorphous states. S. Kohara, K. Kato, S. Kimura, H. Tanaka, Appl. Phys. Lett. 2006, 89, 201910

Limitation: Spontaneous crystallization at ~150 degrees Celsius (423 K)

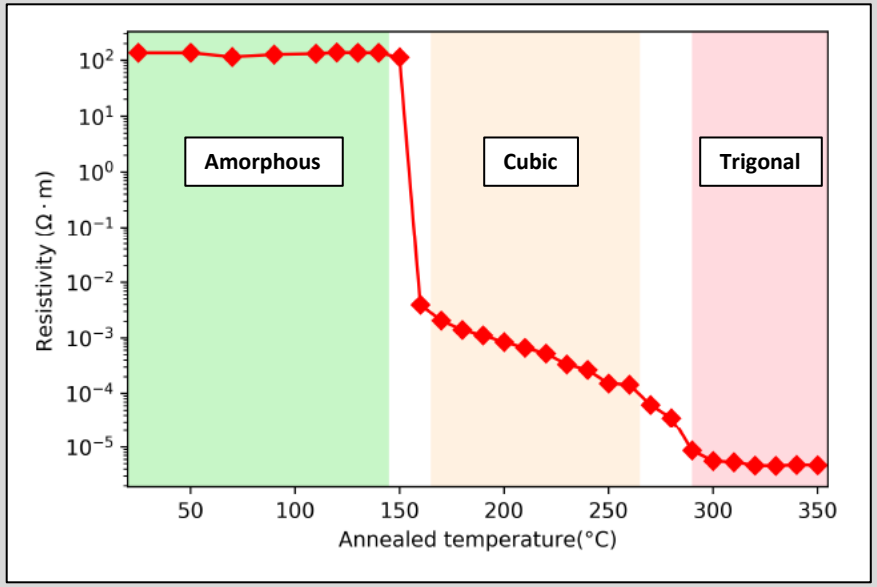
GeSbTe (GST) Alloys

Most prevalent class of PCM due to its fast and reversible phase transitions between amorphous and crystalline states.



Synthesis and screening of phase change chalcogenide thin film materials for data storage. S. Guerin, B. Hayden, D. Hewak, C. Vian, ACS Comb. Sci. 2017, 19, 478–491.

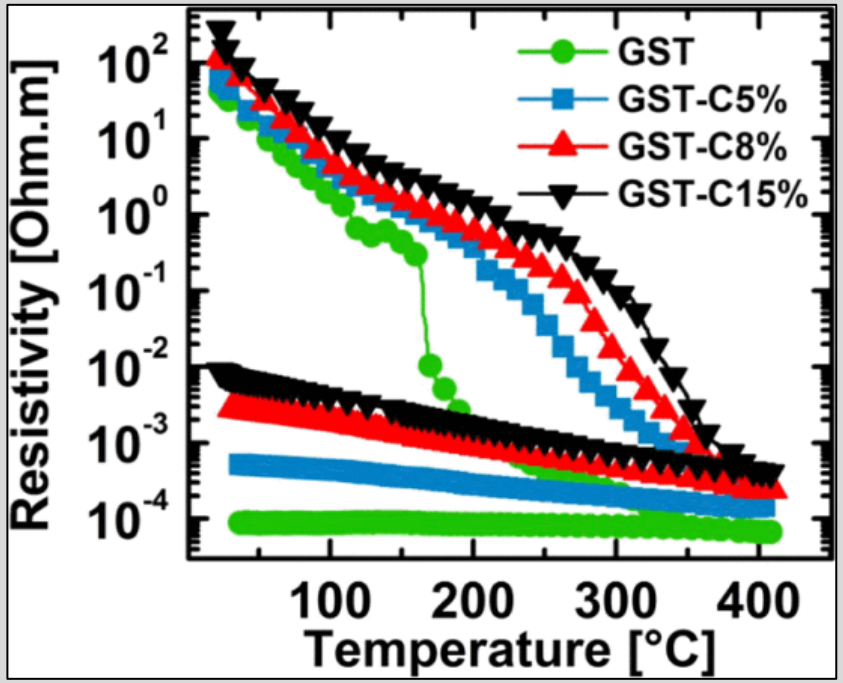
Guo, P.; Sarangan, A.M.; Agha, I. A Review of Germanium-Antimony-Telluride Phase Change Materials for Non-Volatile Memories and Optical Modulators. Appl. Sci. 2019, 9, 530. https://doi.org/10.3390/app9030530



Limitation: Spontaneous crystallization at ~150 degrees Celsius (423 K)

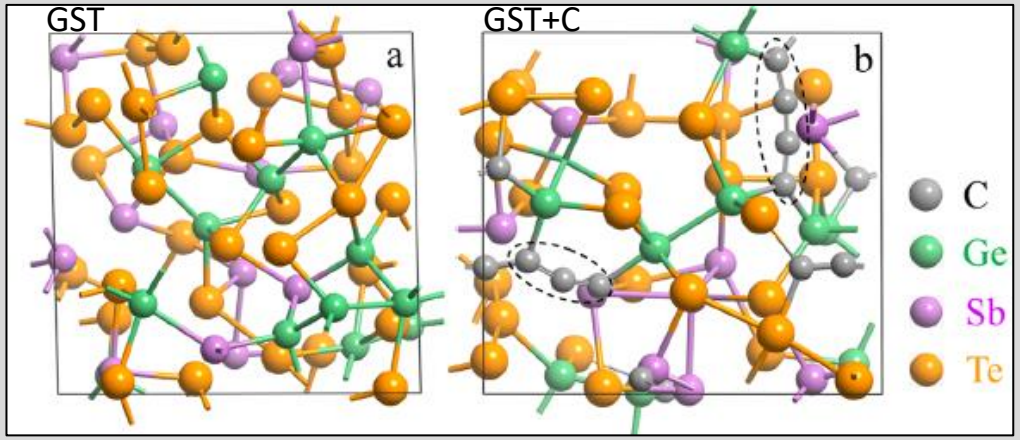
Carbon Doping in GST Systems

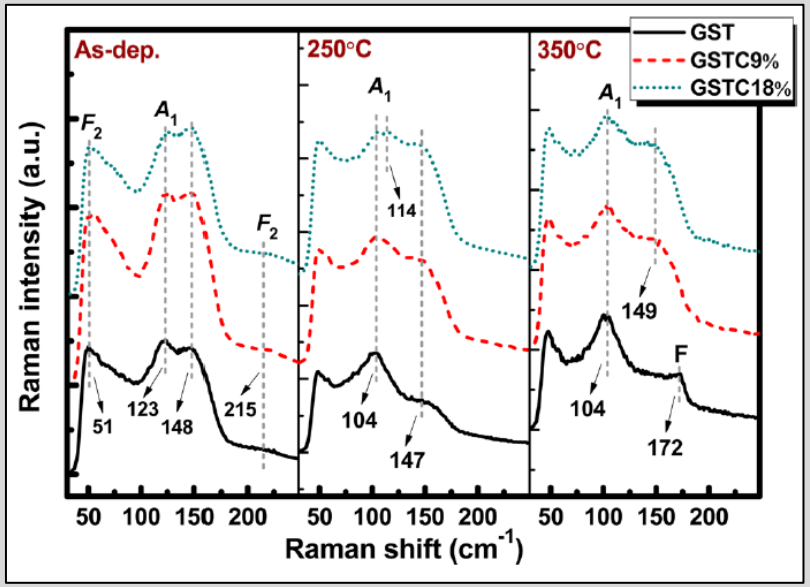
Adding carbon increases the stability of the amorphous phase



Lowering the Reset Current and Power Consumption of Phase-Change Memories with Carbon-Doped Ge2Sb2Te5, Q. Hubert et al, 2012 4th IEEE International Memory Workshop, 2012, pp. 1-4, doi: 10.1109/IMW.2012.6213683.

Understanding Phase-Change Behaviors of Carbon-Doped Ge2Sb2Te5 for Phase-Change Memory Application, X. Zhou et al, ACS Applied Materials & Interfaces (2014)





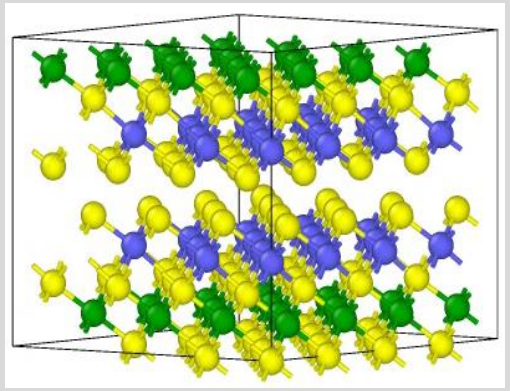
Increased disorder in the amorphous phase
Also stabilizes cubic phase

Current Work: Atomic Simulations

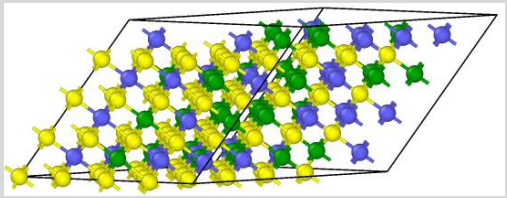
Investigate the structure and stability of amorphous and crystalline $Ge_2Sb_2Te_5$, $(Ge_2Sb_2Te_5)_xC_{1-x}$ (x: 0.9 to 0.99)

MD simulations of each phase for 20ps

Hex-GST 300K

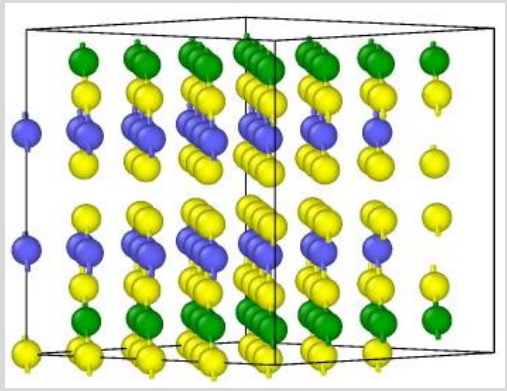


Cub-GST 300K

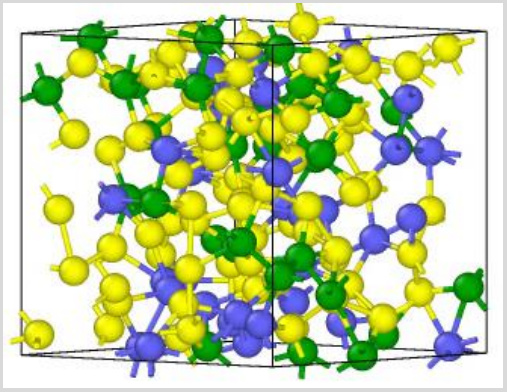


Ge Sb
Te C

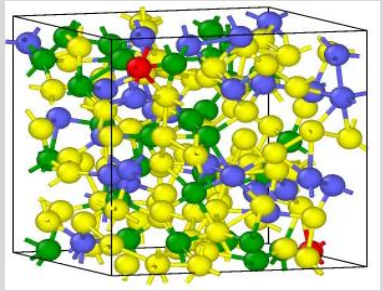
Liq-GST 2000K



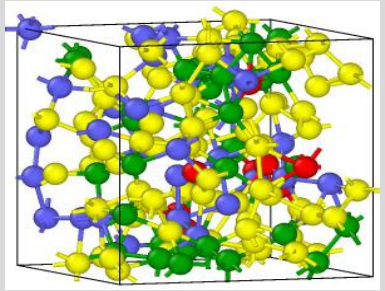
Am-GST 300K



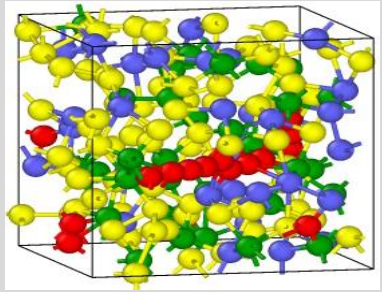
Am-GST+1%C 300K



Am-GST+5%C 300K



Am-GST+10%C 300K



Hexagonal Lattice Params:

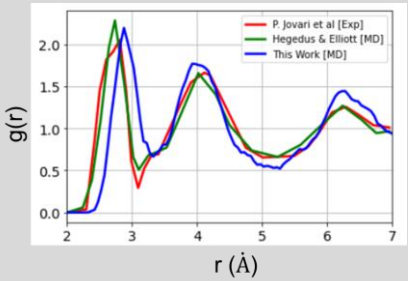
a_{exp} = 4.22 Å || c_{exp} = 17.18 Å

a_{calc} = 4.23 Å || c_{calc} = 17.15 Å

Cubic-Rocksalt Lattice Params:

a_{exp} = 6.02 Å

a_{calc} = 6.00 Å



Jovari, P. et al. 'Wrong bonds' in sputtered amorphous Ge2Sb2Te5 . *J. Phys. Condens. Matter* 19, 335212 (2007).

Hegedüs, J., Elliott, S. Microscopic origin of the fast crystallization ability of Ge–Sb–Te phase-change memory materials. *Nature Mater* 7, 399–405 (2008). https://doi.org/10.1038/nmat2157

Explore phase stabilities via Free Energy

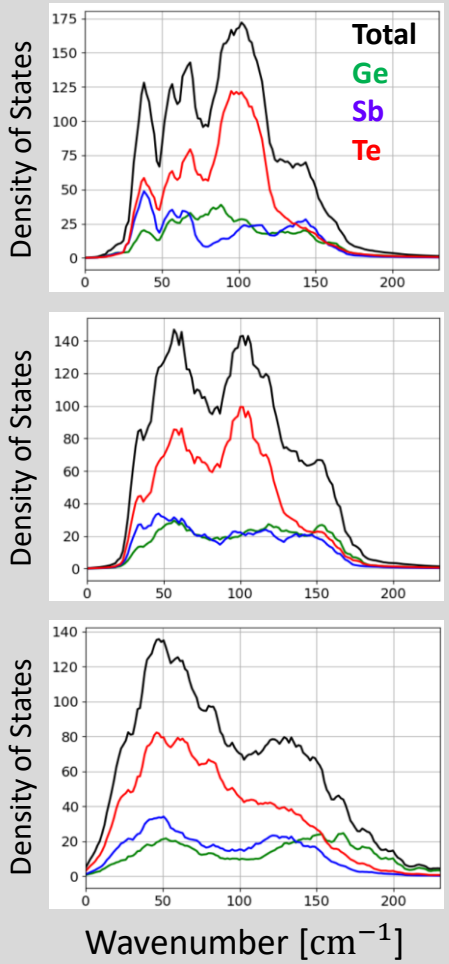
Investigate the structure and stability of amorphous and crystalline $Ge_2Sb_2Te_5$, $(Ge_2Sb_2Te_5)_xC_{1-x}$ (x: 0.9 to 0.99)

GST 225

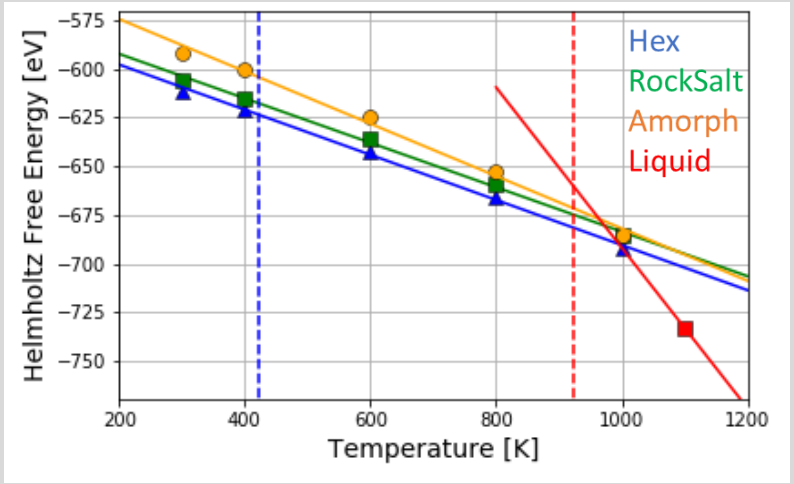
Hex

Cub

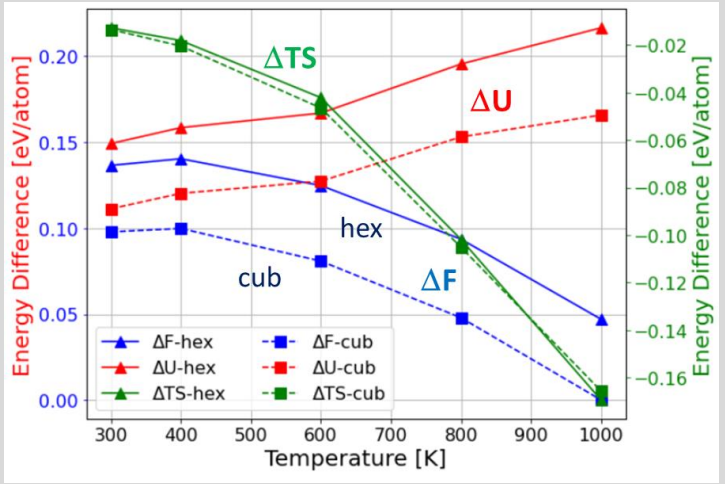
Am



GST225: Free Energy Landscape



GST225: Hex & Cubic vs Amorph



The cub-GST phase is meta stable and reduced difference in the free energy relative to the amorph phase is dominated by contributions from internal energy

Explore phase stabilities via Free Energy

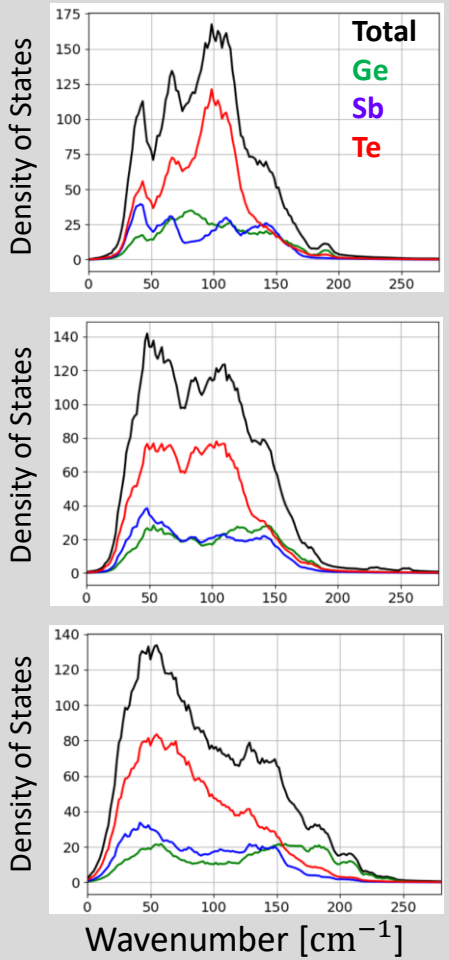
Investigate the structure and stability of amorphous and crystalline $Ge_2Sb_2Te_5$, $(Ge_2Sb_2Te_5)_xC_{1-x}$ (x: 0.9 to 0.99)

GST + 1%C

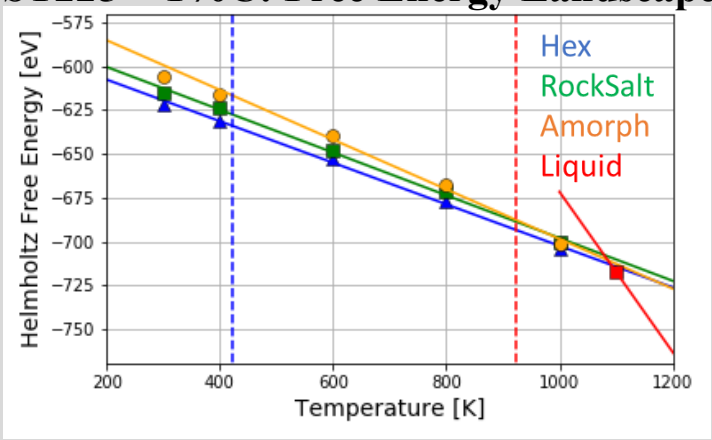
Hex

Cub

Am

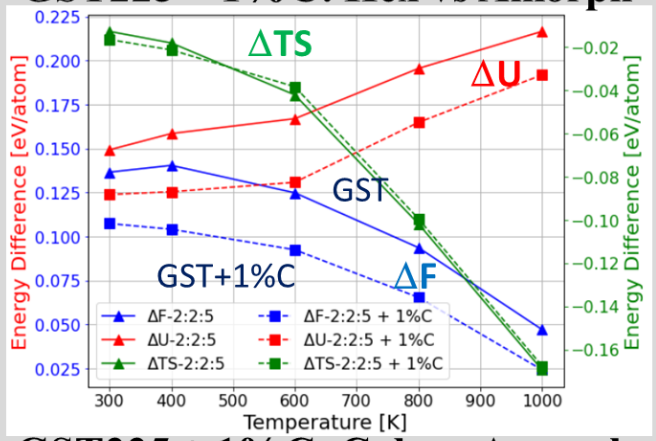


GST225 + 1%C: Free Energy Landscape

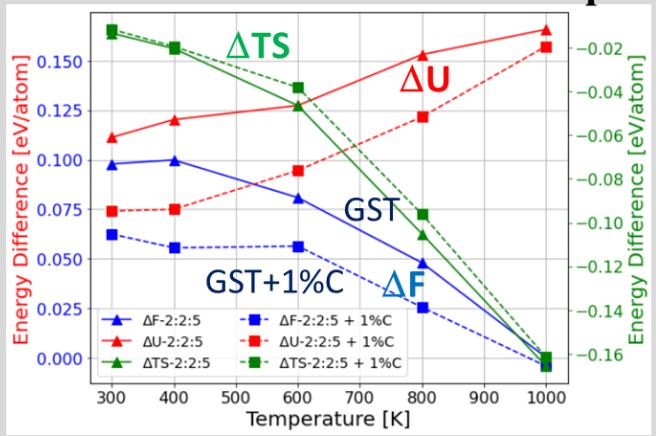


Adding 1% carbon stabilizes the amorphous phase by shifting internal energy

GST225 + 1%C: Hex vs Amorph

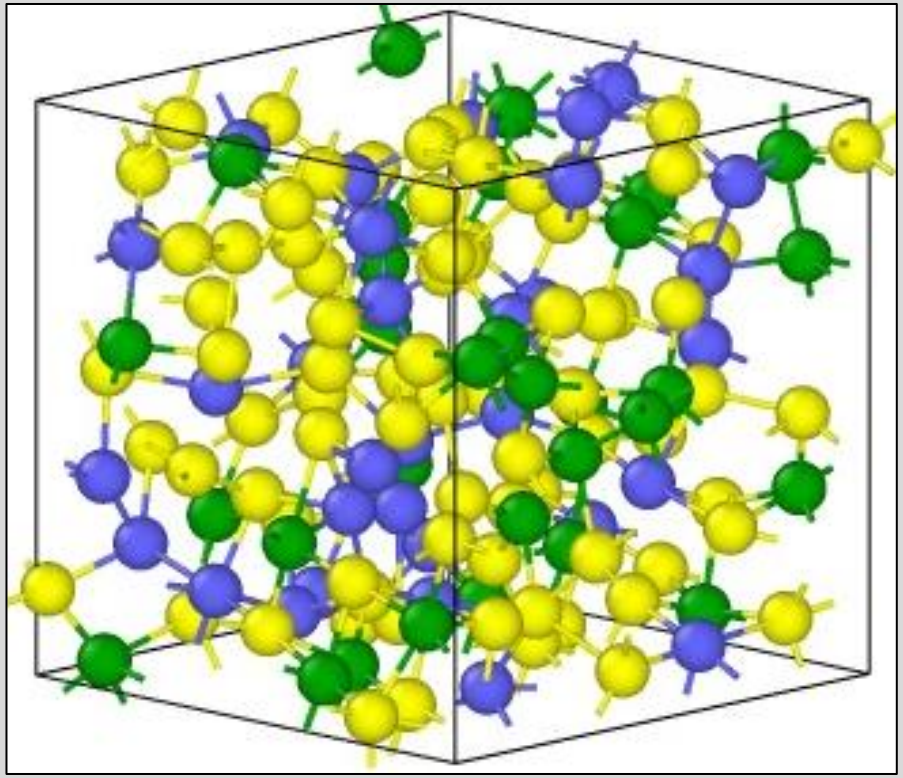


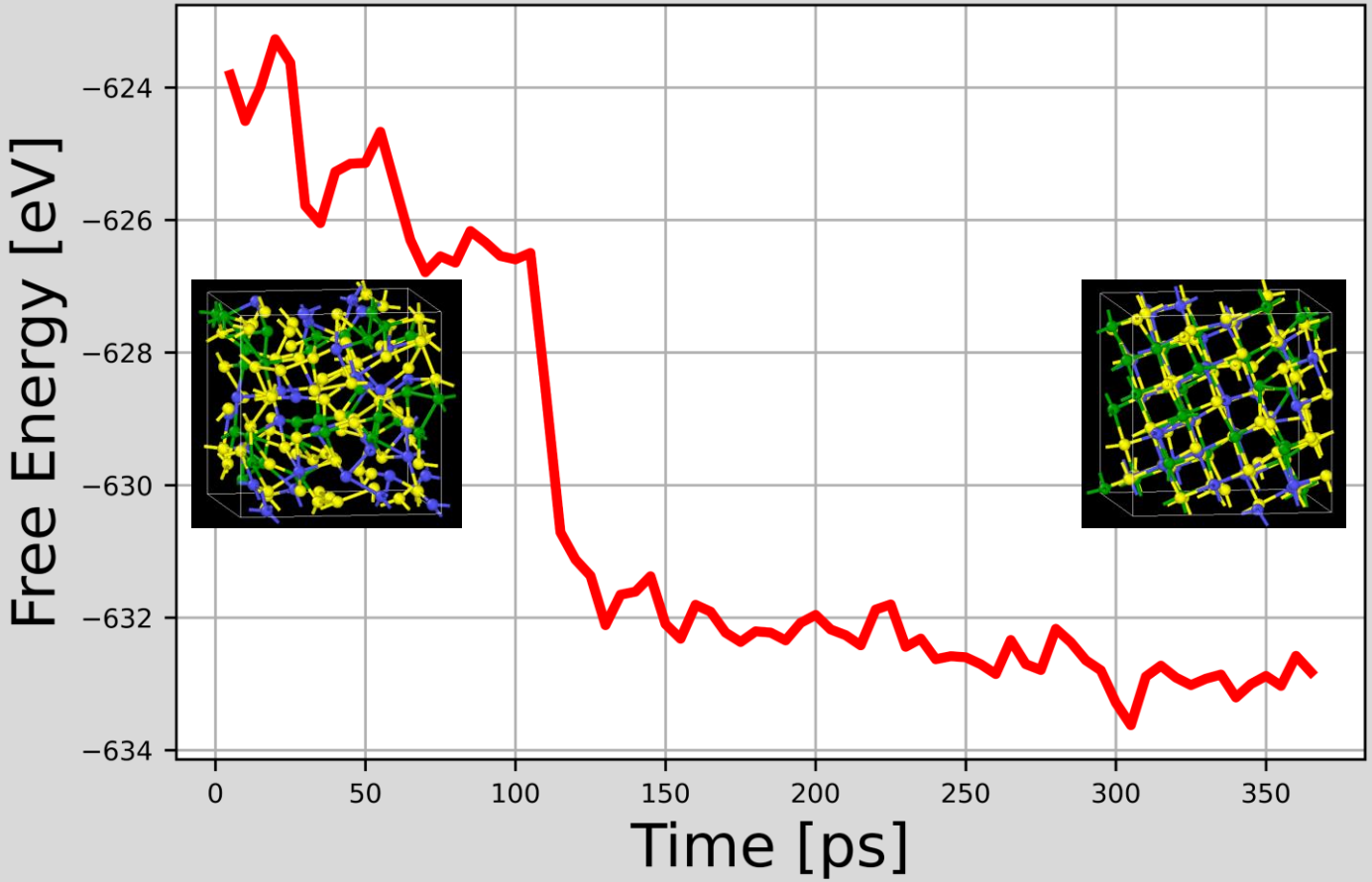
GST225 + 1%C: Cub vs Amorph



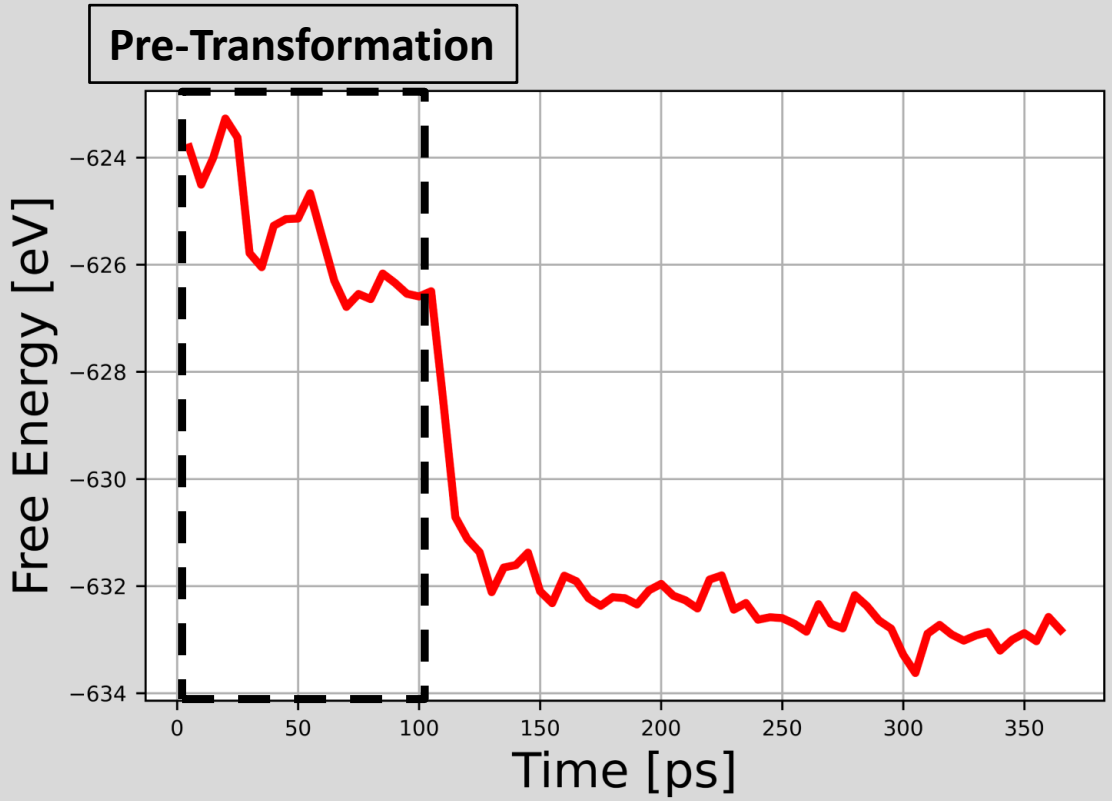
Crystallization via molecular dynamics

MD Simulation @ 600K

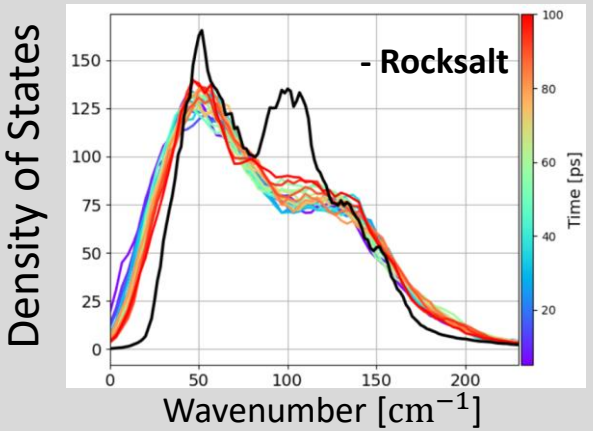


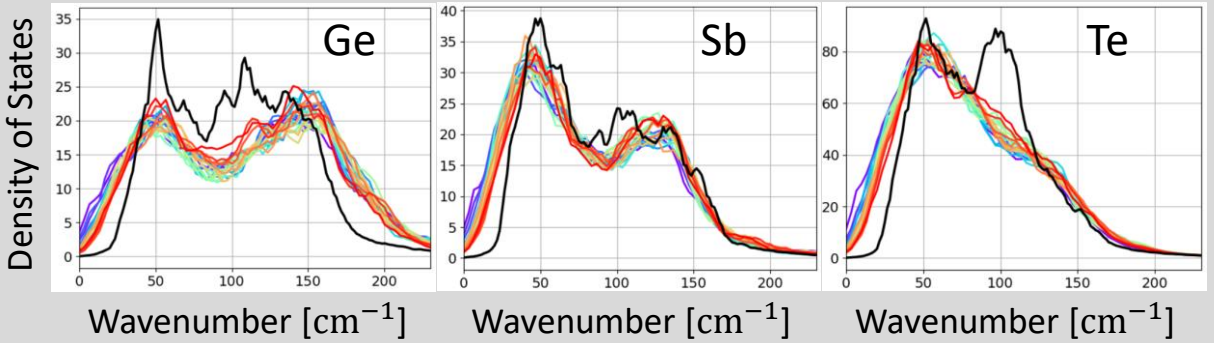


Crystallization via molecular dynamics



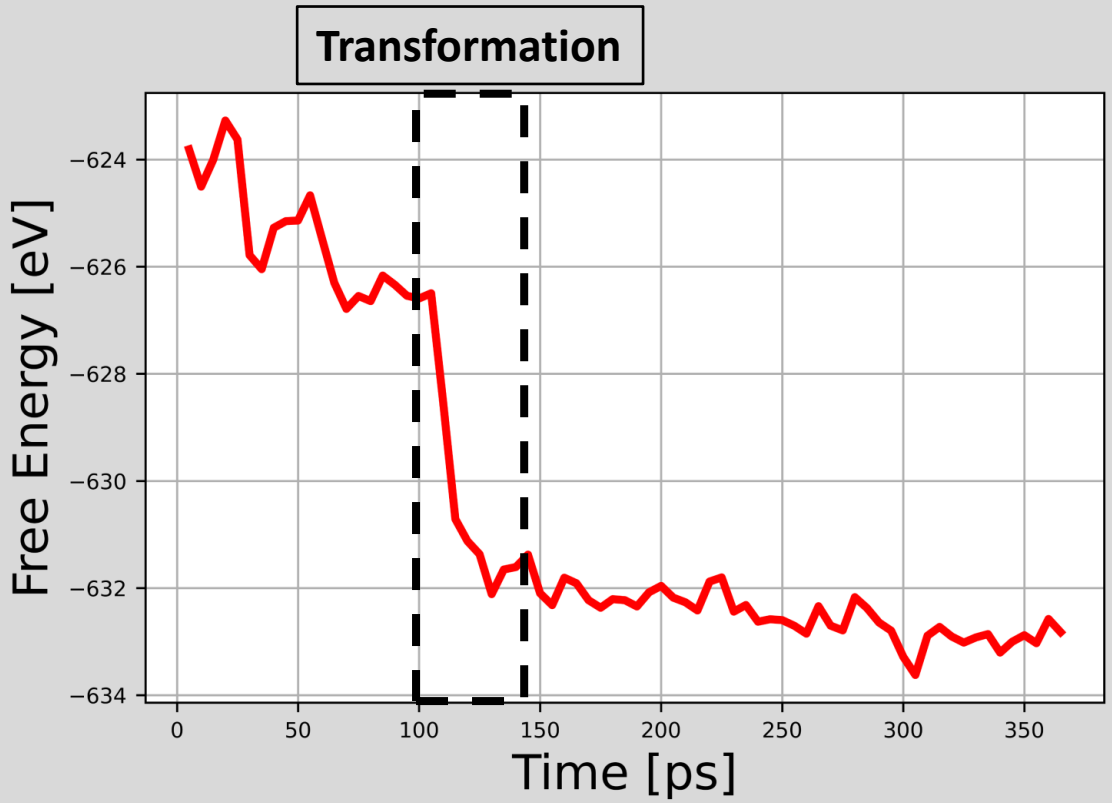
Evolution of Vibration DoS from 5ps – 100ps



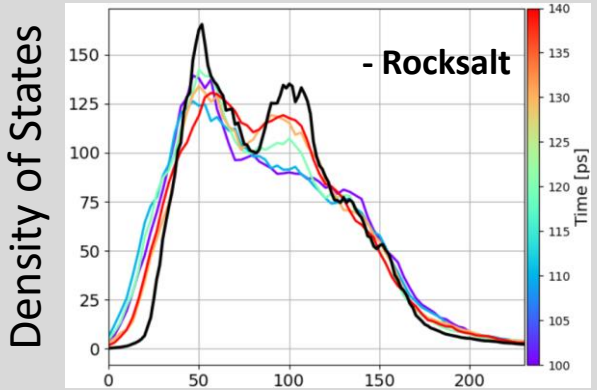


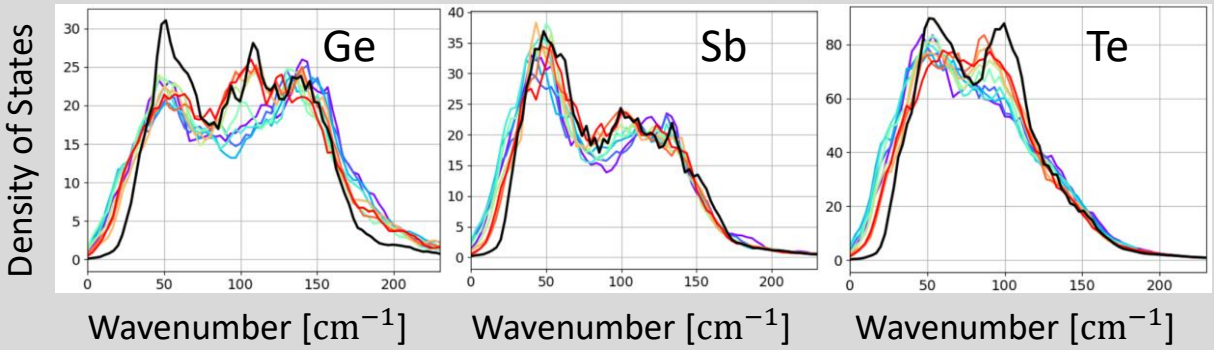
Much larger difference in spectra for Ge and Te than for Sb

Crystallization via molecular dynamics



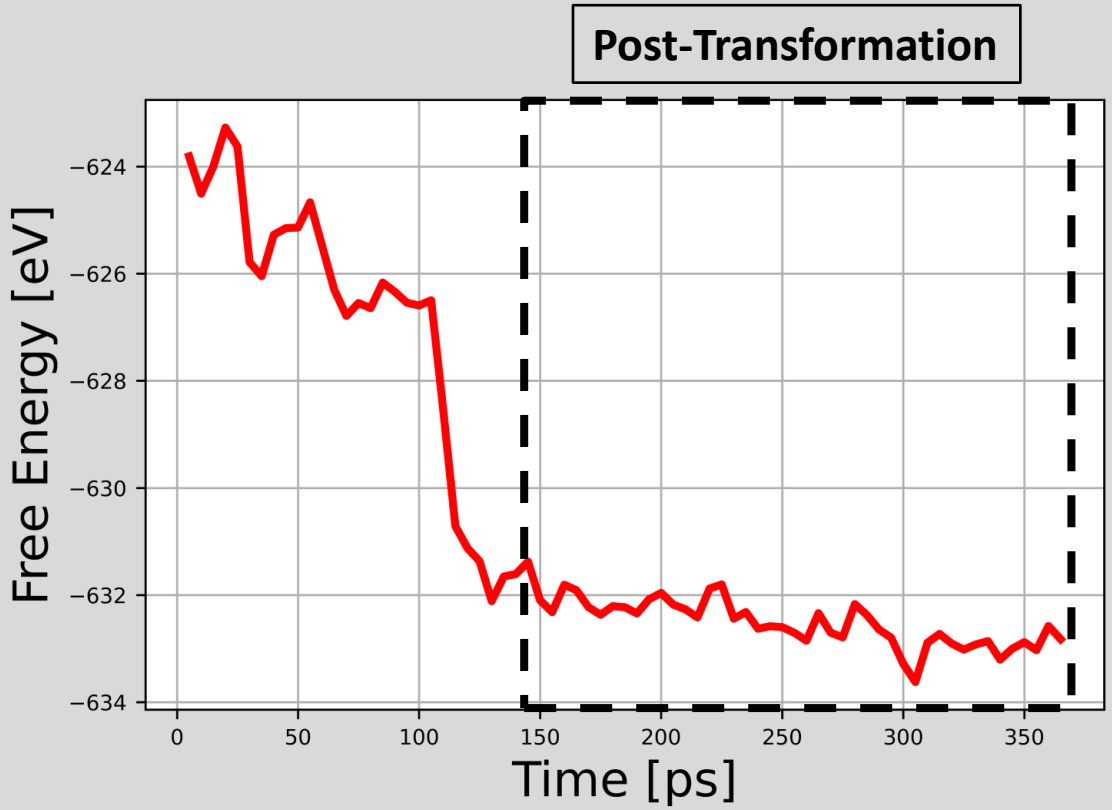
Evolution of Vibration DoS from 100ps – 140ps



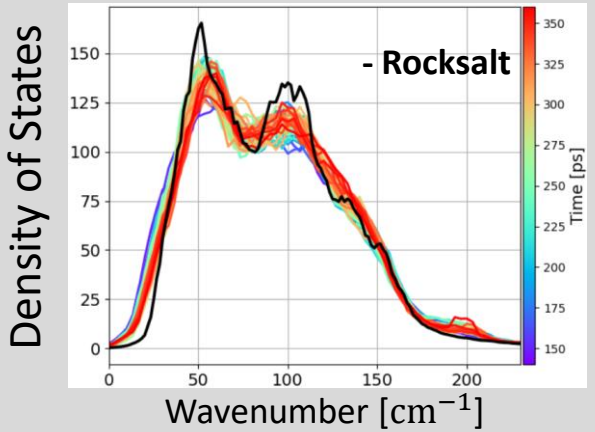


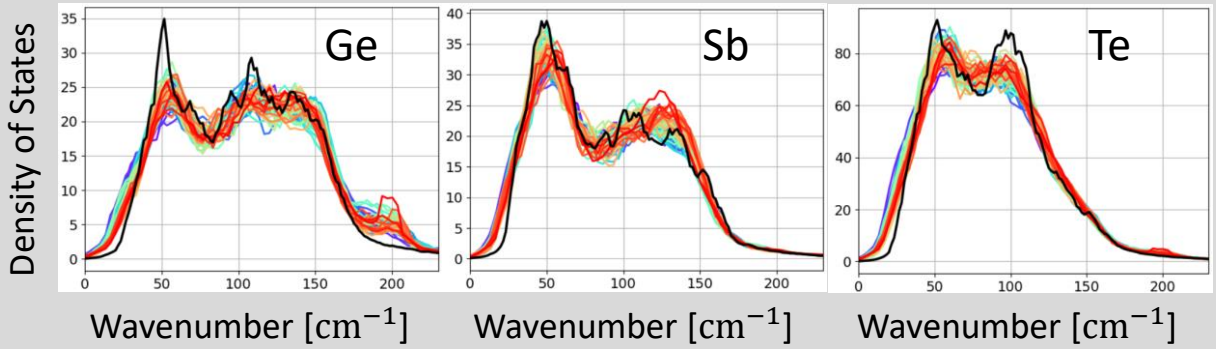
Local bonding environments of the Ge-Te network show the most change during crystallization

Crystallization via molecular dynamics



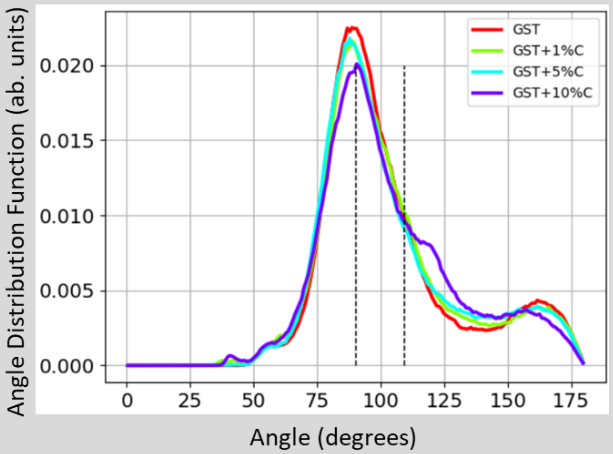
Evolution of Vibration DoS from 140ps – 360ps

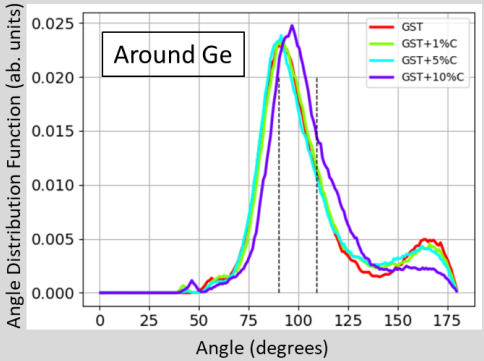


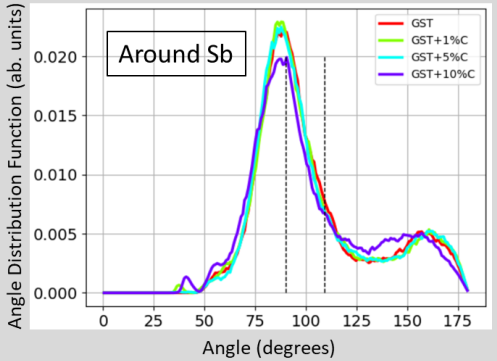


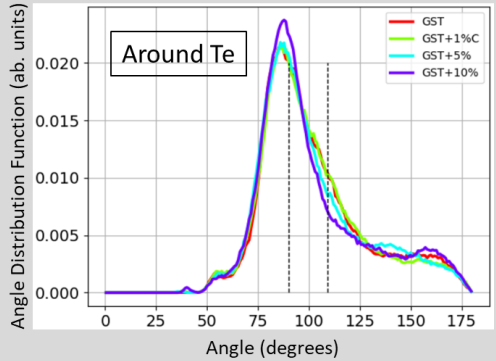
Spectra for Ge and Te continue to evolve while Sb is steady

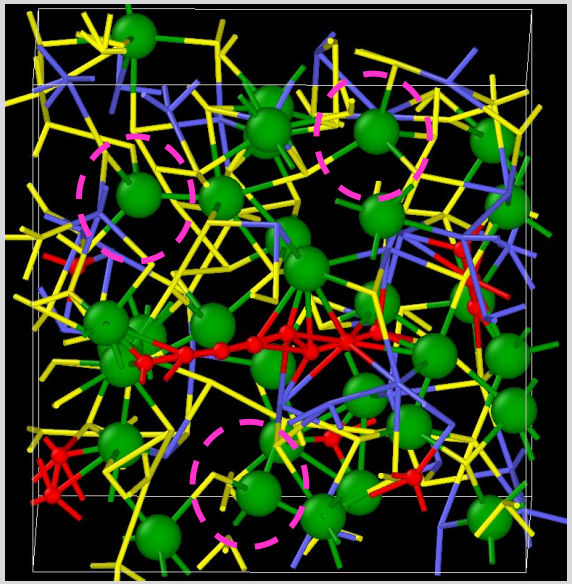
How does carbon change the amorphous phase?

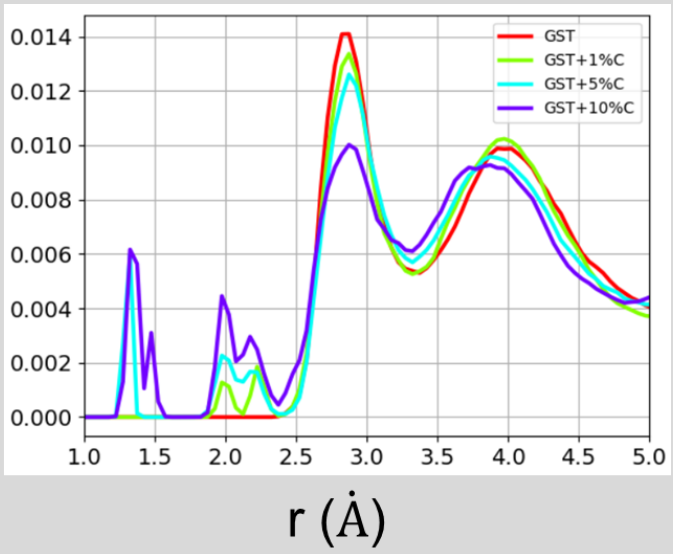


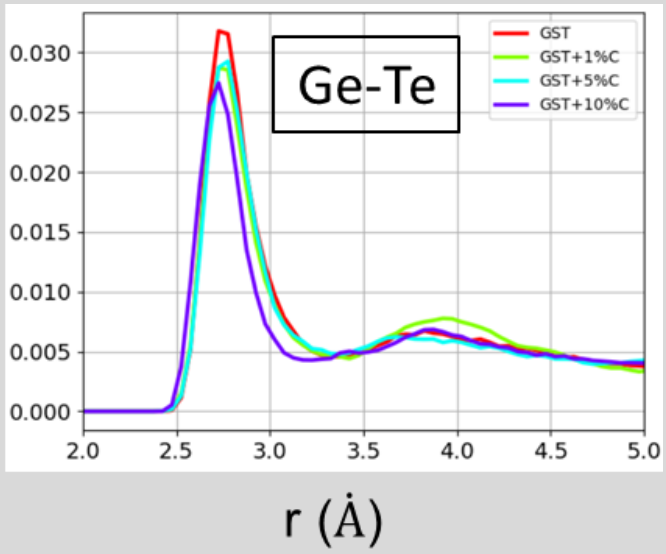




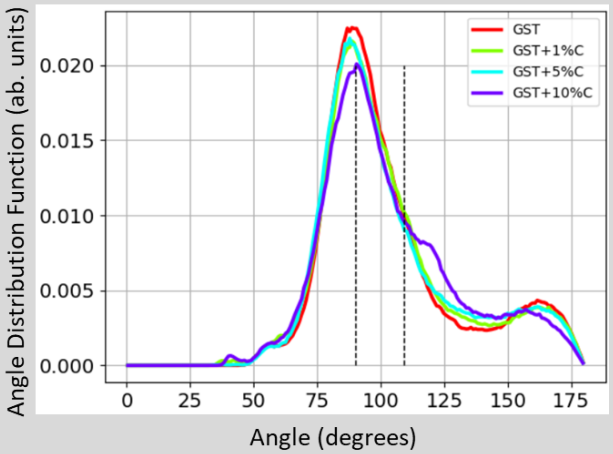




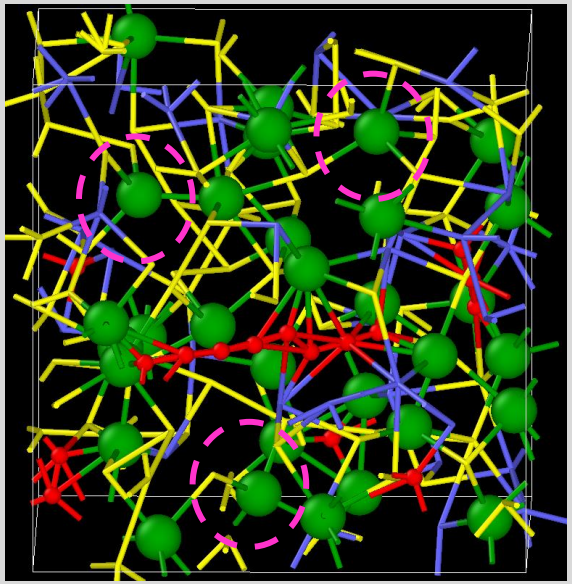


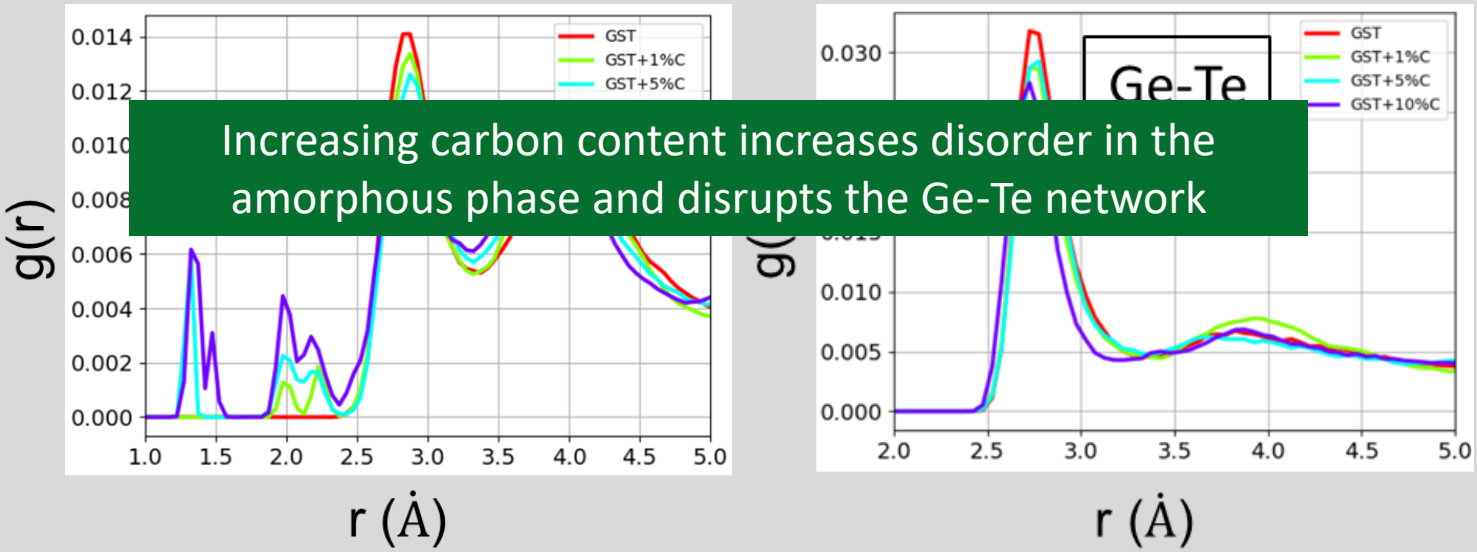


How does carbon change the amorphous phase?



Increasing carbon content alters the local environment of Ge atoms resulting in more tetrahedral bonding





Increasing carbon content increases disorder in the amorphous phase and disrupts the Ge-Te network

Final Remarks

Conclusions

- Adding carbon to GST decreases the free energy difference between the crystalline and amorphous phases by shifting the contribution from the internal energy.
- During crystallization of undoped GST, most of the changes to local bonding environments are due to the evolution of the Ge-Te network.
- Analysis of the doped and undoped amorphous phases suggests that increased carbon content leads to
  - A decrease in overall octahedral bonding
  - An increase in tetrahedral bonded Ge atoms
  - Higher disorder
  - Disruption of the Ge-Te network.

Future Work

- Vibrational analysis of amorphous carbon doped GST structures.
- Further analysis of the evolution of atomic structure during crystallization of undoped GST.
- Crystallization of carbon doped GST systems.
- Investigation of off-stoichiometric doped and undoped GST systems (i.e., Ge-rich and Ge-poor).

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