Pressure Induced Amorphization and Grain Segmentation in $Ge_2Sb_2Te_5$

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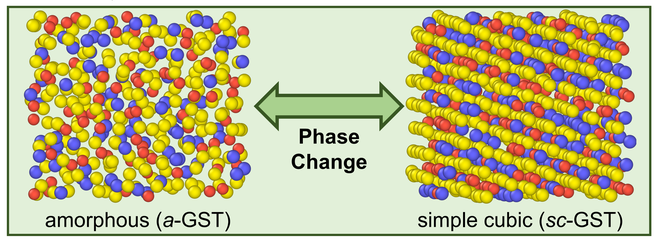
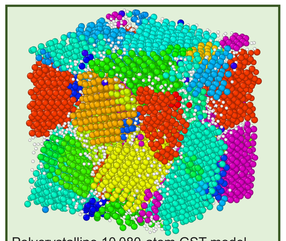
Phase Change Materials

$Ge_2Sb_2Te_5$ (GST) is a phase change material known for its ability to rapidly switch between its amorphous and crystalline states. Both **states are stable and exhibit distinct optical and electronic properties,** making them key candidates for non-volatile, high-speed data memory applications.



In the crystalline state, GST forms a **polycrystalline structure** made of multiple grains with varying orientations and separated with grain boundaries. These factors affect the properties of these materials, making them important to investigate.

Recent advancements in computational methods make computer simulations of large-scale polycrystalline systems, enabling studies of its structure.

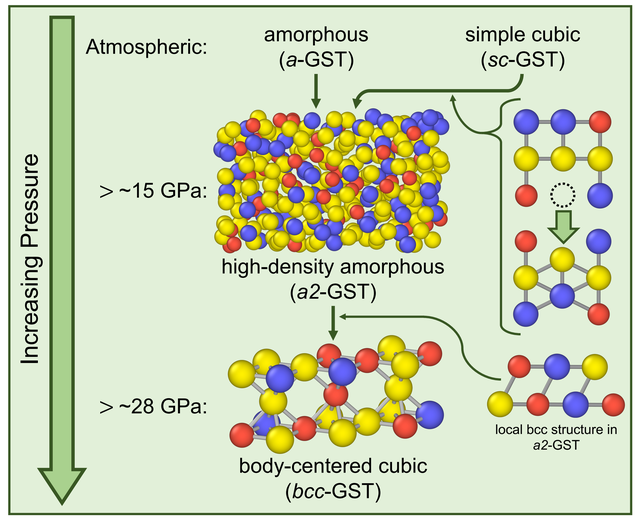


Polycrystalline 10 080-atom GST model. Colours indicate individual crystal grains and white atoms are grain boundaries.

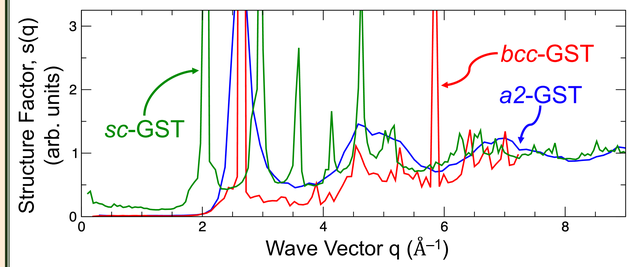
High Pressure Behaviour[1]

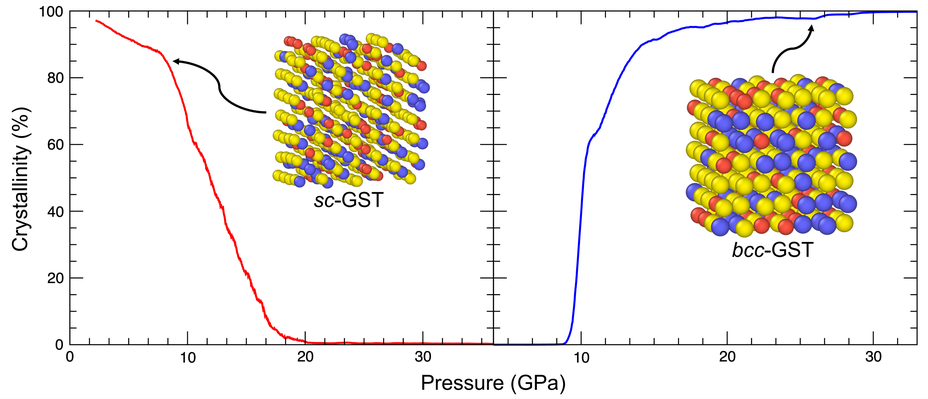
Previous experimental and computational studies have shown phase transitions of GST at high pressures. Existing computational simulations have only occurred in small systems (< 300 atoms).

These transitions are **driven by the elimination of vacancies** in its structure, when subjected to high pressures.



Pressure-Induced Transitions





Using the new ACE potential, GST can be simulated to higher pressures. When compressing *sc*-GST, it transitions to *a2*-GST, evident in both the **crystallinity and structure factor.**

Although *bcc*-GST can not be formed via compression (likely due to the long-time scales needed), the structure is stable and an amorphization is observed upon decompression.

Methods

Crystal Identification[2]

1. Neighbour lists ($\mathcal{N}(i)$) of each atom are created containing all atoms within a distance r_c.
   - r_c is determined by the first peak of the radial distribution function
2. A **local orientational order parameter** for each atom is calculated using spherical harmonics:

$$\bar{q}_{\ell,m}(i) = \frac{1}{|\mathcal{N}(i)|} \sum_{j \in \mathcal{N}(i)} Y_{\ell,m}(\hat{\mathbf{r}}_{ij})$$

   - $\ell = 4$ is used for simple cubic structures
   - $\ell = 6$ is used for body-centered cubic structures
3. The **local environment** of each atom is described by:

$$\mathbf{q}_\ell(i) = \frac{1}{\sqrt{\sum_{m=-\ell}^{\ell} |\bar{q}_{\ell,m}(i)|^2}} \begin{pmatrix} \bar{q}_{\ell,-\ell}(i) \\ \vdots \\ \bar{q}_{\ell,\ell}(i) \end{pmatrix}$$

4. Atom $\boldsymbol{i}$ is "connected" to atom $\boldsymbol{j}$ if:
$$\Re(\mathbf{q}_\ell(i) \cdot \mathbf{q}_\ell(j)) > 0.75$$
5. Atom $\boldsymbol{i}$ is **classified as crystalline** if it has 3 or more connected neighbours.

Simulation

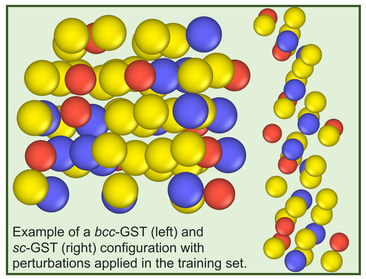
The simulations were performed with LAMMPS Molecular Dynamics Simulator[3] with machine-learned (ML) **Atomic Cluster Expansion (ACE) potentials**.[4] These potentials provide considerable computational speed, while remaining highly accurate.

As **no existing ML potential** can model GST at high-pressure, a new ACE potential was created.

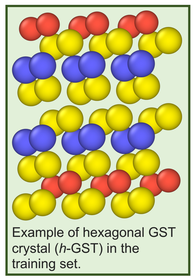
It was trained using a published dataset combined with new high-pressure configurations.

System	Configurations	Atoms
Published dataset[5]	4 520	469 194
sc-GST [a]	204	11 016
h-GST*, [a]	204	11 016
a-GST [a]	400	21 600
bcc-GST [b]	750	54 000
Total	**6 078**	**566 826**

*hexagonal GST [a] ~7.5–20 GPa [b] ~15–33 GPa



Example of a *bcc*-GST (left) and *sc*-GST (right) configuration with perturbations applied in the training set.



Example of hexagonal GST crystal (*h*-GST) in the training set.

Crystal Grains

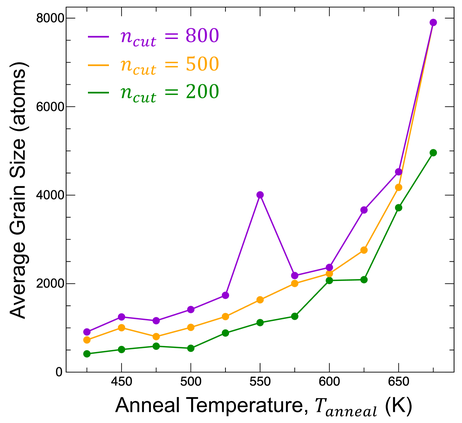
The following algorithm was created to sort the crystalline atoms into individual crystal grains:

1. Atom $\boldsymbol{i}$ is classified as an **"inner" crystalline atoms** if it has 5 or more crystalline neighbours
2. Inner crystalline atoms are grouped into **crystal grains**:
   - Atom $\boldsymbol{i}$ is in the same grain with atom $\boldsymbol{j}$, if:
$$R(\boldsymbol{q}_\ell(i) \cdot \boldsymbol{q}_\ell(j)) > 0.97, \quad j \in N(i)$$
3. Transient grains containing fewer than 4 atoms are removed
4. Remaining crystalline atoms are grouped into **existing crystal grains**:
   - Atom $\boldsymbol{i}$ is matched to the same grain as atom $\boldsymbol{j}$ which:
$$\max_{j \in N(i)} [\Re(\mathbf{q}_\ell(i) \cdot \mathbf{q}_\ell(j))] > 0.80$$
5. Step 4 is repeated until no new crystalline atoms are placed into grains

A published ACE potential[6] was used to model the 10 080-atom GST systems.

***a*-GST was heated from 300 K to a target temperature T_{anneal}** over 200 ps and held at T_{anneal} for 2 ns, all under atmospheric pressure.

The average grain size was plotted for each T_{anneal}. To focus on the dominant grains, **a cutoff n_{cut} sets a minimum size for the grains** included in the average. This avoids skewing due to the large number of small grains.



Future Work

Pressure-induced amorphization:

- Determine the regimes where the *bcc*-GST and *a*2-GST dominate
- Use Voronoi analysis to study changes in vacancies upon compression

Grain segmentation:

- Characterize grain structure in crystals prepared using different methods
- Analyze the kinetics along grain boundaries
- Explore grain boundaries as sites of amorphous nucleation
- Determine the effect of the grain structure on its electrical properties

Acknowledgements

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References

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[3] A.P. Thompson et al., "LAMMPS - a flexible simulation tool for particle-based materials modeling at the atomic, meso, and continuum scales," Computer Physics Communications **271**, 108171 (2022).

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[6] O.R. Dunton, T. Arbaugh, and F.W. Starr, "Computationally efficient machine-learned model for GST phase change materials via direct and indirect learning," The Journal of Chemical Physics 162(3), (2025).