

**Density functional simulations of decomposition pathways of Ge-rich GeSbTe alloys for phase  
change memories  
Supplemental Material**

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## I. CRYSTALLINE PHASES

We give here details on the construction of the crystalline models of the different alloys.

In the case of GST523, GST423 and GST221 there are less than three  $p$  electrons per atom in the formula unit and thus cationic vacancies have not been included while antimony is supposed to behave as an amphoteric element due to the low content of Te. In GST523, the cation sublattice is fully occupied by Ge atoms while the anionic sublattice is occupied by the Sb and Te atoms. Special quasi-random structures (SQS)<sup>1</sup> models were generated with the anionic sublattice occupied by 64 atoms of Te and 44 atoms of Sb which corresponds to an actual composition of  $\text{Ge}_{4.909}\text{Sb}_2\text{Te}_{2.909}$ .

The GST423 alloy is modeled instead with four Ge atoms and 0.5 Sb atoms per formula unit on the cation sublattice and three Te atoms and 1.5 Sb atoms per formula unit on the anionic sublattice. SQS models were generated with the cationic sublattice occupied by 96 atoms of Ge and 12 atoms of Sb while the anionic sublattice is occupied by 36 atoms of Sb and 72 atoms of Te which correspond to the exact composition GST423.

The GST323 alloy has exactly three  $p$  electrons per site, the cationic sublattice is occupied by three Ge atoms and one Sb atom per formula unit, while the anionic sublattice is occupied by three Te atoms and one Sb atom per formula unit. SQS models were generated with the cationic sublattice occupied by 81 atoms of Ge and 27 atoms of Sb while the anionic sublattice is occupied by 81 atoms of Te and 27 atoms of Sb which correspond to the exact composition GST323.

The GST223 and GST123 alloys have more than three  $p$  electrons per atom in the formula unit and then we introduced vacancies in the cationic sublattice to enforce the presence of an average of three  $p$  electrons per site in the rocksalt crystal. SQS models of GST223 were generated with the cationic sublattice occupied by 60 atoms of Ge, 39 atoms of Sb and 9 vacancies while the anionic sublattice is occupied by 87 atoms of Te and 21 atoms of Sb which corresponds to the composition  $\text{Ge}_2\text{Sb}_2\text{Te}_{2.900}$ . SQS models of GST123 were generated with the cationic sublattice occupied by 33 atoms of Ge, 54 atoms of Sb and 21 vacancies while the anionic sublattice is occupied by 96 atoms of Te and 12 atoms of Sb which corresponds to the composition  $\text{Ge}_1\text{Sb}_2\text{Te}_{2.909}$ .

Finally, the GST221 alloy was described by SQS models with the cationic sublattice occupied by 86 atoms of Ge and 22 atoms of Sb while the anionic sublattice is occupied by 64 atoms of Sb and 44 atoms of Te which corresponds to the composition  $\text{Ge}_2\text{Sb}_2\text{Te}_{1.02}$ .

## II. ENERGETICS OF DECOMPOSITION REACTIONS IN THE CRYSTALLINE PHASES

In order to estimate the reaction energies, we also included the configurational entropy in the crystalline phases due to disorder as given by  $S = \frac{-k_B}{2} \sum_{i,j} x_{i,j} \ln(x_{i,j})$  where the  $j$  index runs over the sublattices (i.e. cationic and anionic) and the  $i$  index runs over the atomic species which occupy a particular sublattice;  $k_B$  is the Boltzmann constant and  $x_{i,j}$  is the molar fraction. The factor two at the denominator is due to the fact that we are summing the contribution to the entropy of two sublattices. For example, the entropy of mixing of GST221, which was modeled with a composition of  $\text{Ge}_{0.8}/\text{Sb}_{0.2}$  in the cationic sublattice and a composition of  $\text{Sb}_{0.6}/\text{Te}_{0.4}$  in the anionic one, was calculated as  $S = \frac{-k_B}{2} (0.2 \cdot \ln(0.2) + 0.8 \cdot \ln(0.8) + 0.6 \cdot \ln(0.6) + 0.4 \cdot \ln(0.4))$ .

To compute the reaction energy, we also needed the reference energy of Ge, GeTe and  $\text{Sb}_2\text{Te}_3$  crystalline phases. Because of the non-perfect stoichiometry of some of our models due to the constraint imposed by the 216-atom supercell, the reference energy of crystalline Sb is needed as well.

A 216-atom supercell was used to model crystalline Ge in the diamond structure and crystalline Sb in the  $R\bar{3}m$  structure<sup>2</sup> with BZ integration restricted to the supercell  $\Gamma$  point. The resulting equilibrium lattice parameter of Ge of 5.771 Å is very close to the value of 5.751 Å obtained at full convergence in the BZ

integration with the Quantum-Espresso (QE) suite of programs which employs a plane wave expansion of KS orbitals<sup>3</sup>. In all benchmark calculations with the QE code, we used the same pseudopotentials employed with the CP2k code. The resulting equilibrium lattice parameters of Sb in turn are  $a=4.503$  Å and  $\alpha=59.56^\circ$  which are very close to the experimental values of  $a=4.507$  Å and  $\alpha=57.11^\circ$  from Ref. 2. Crystalline  $\alpha$ -GeTe in the  $R\bar{3}m$  structure<sup>4</sup> was modeled in a 162 atoms supercell (i.e.  $3\times 3\times 3$  trigonal cells). The resulting lattice parameters  $a=4.36$  Å and  $\alpha=59.16^\circ$  are close to the experimental values of  $a=4.31$  Å and  $\alpha=57.9^\circ$  from Ref. 4. Crystalline  $\text{Sb}_2\text{Te}_3$  ( $R\bar{3}m$  space group)<sup>5</sup> was modeled in a 270-atom supercell ( $3\times 3\times 2$  conventional hexagonal 15-atom unit cells). The resulting lattice parameters  $a=4.387$  Å and  $c=29.550$  Å are in good agreement with the experimental values of  $a=4.264$  Å and  $c=30.458$  Å from Ref. 5.

To compute the reaction free energy, the total energy of all compounds was then computed with a  $3\times 3\times 3$   $k$  point mesh in the supercell Brillouin zone. The energy and entropy per atom of the different alloys or reference systems are collected in Table S1. The energies of the amorphous models discussed in Sec. IIIB in the article are also reported in Table S1 for the sake of completeness.

TABLE S1: Total energy of the different alloys in the crystalline and amorphous phases. The ideal entropy of mixing for the crystalline phase is also given. The error bar is obtained by averaging over three different SQS models. Information on the amorphous models is given in Sec. IIIB in the article. Two amorphous models of GST523 were generated (M1 and M2) to assess the variability of the total energy on models generated independently by quenching from the melt.

Compound	Energy (eV/atom)	S (meV/K/atom)	S (meV/K/f.u.)
Ge	-107.1350	-	-
Sb	-150.8936	-	-
GeTe	-164.8953	-	-
GeTe (amorphous)	-164.7736	-	-
$\text{Sb}_2\text{Te}_3$	-193.9511	-	-
$\text{Ge}_1\text{Sb}_2\text{Te}_{2.909}$	-178.749±0.004	0.05837	0.3891
$\text{Ge}_2\text{Sb}_2\text{Te}_{2.9}$	-168.325±0.002	0.05992	0.4394
$\text{Ge}_3\text{Sb}_2\text{Te}_3$	-161.366±0.001	0.04845	0.3876
$\text{Ge}_3\text{Sb}_2\text{Te}_3$ (amorphous)	-161.2696	-	-
$\text{Ge}_4\text{Sb}_2\text{Te}_3$	-155.3120±0.001	0.04244	0.3820
$\text{Ge}_4\text{Sb}_2\text{Te}_3$ (amorphous)	-155.2332	-	-
$\text{Ge}_{4.9095}\text{Sb}_2\text{Te}_{2.909}$	-150.204±0.003	0.02899	0.2899
$\text{Ge}_{4.9095}\text{Sb}_2\text{Te}_{2.909}$ (amorphous M1)	-150.13984	-	-
$\text{Ge}_{4.9095}\text{Sb}_2\text{Te}_{2.909}$ (amorphous M2)	-150.1334	-	-
$\text{Ge}_2\text{Sb}_2\text{Te}_1$	-148.015±0.001	0.05056	0.2528
$\text{Ge}_2\text{Sb}_2\text{Te}_1$ (amorphous)	-147.9471	-	-

To compute the reaction energy, we have to consider the small deviation from stoichiometry imposed by the use of a 216-atom cell. Therefore reactions (1)-(10) in the article must actually be read as

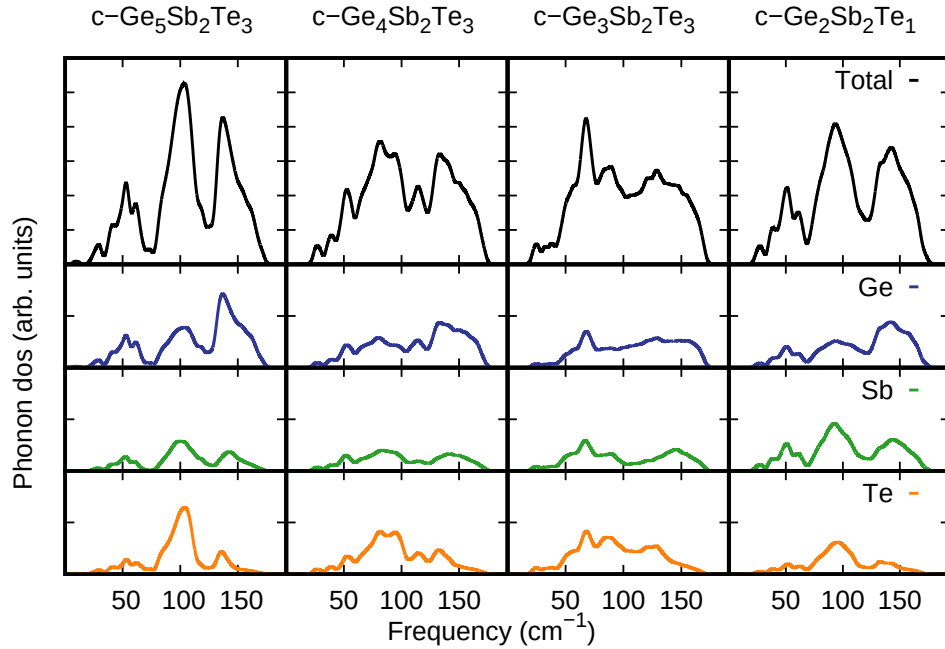
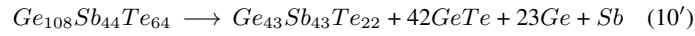
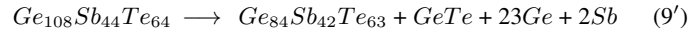
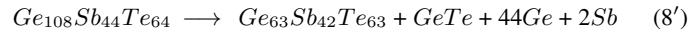
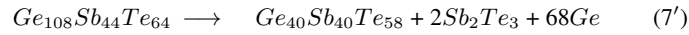
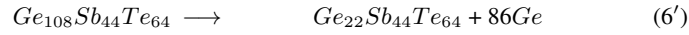
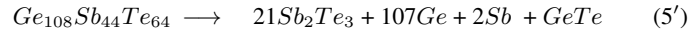
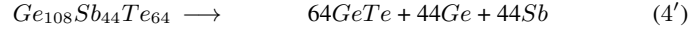
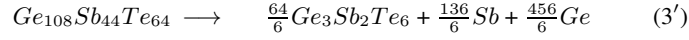
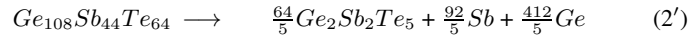
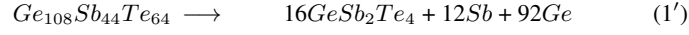


FIG. S1: Phonon density of states of Ge-rich crystalline alloys and their projection on the different types of atoms from DFT phonons at the supercell  $\Gamma$  point broadened by a Gaussian function with a variance of  $1.5 \text{ cm}^{-1}$ . The projections on the different types of atoms are obtained from the eigenvectors of the dynamical matrix (not divided by the square root of the mass) in such a way that the sum of the projected DOS gives the total DOS.

### III. STRUCTURAL, ELECTRONIC AND VIBRATIONAL PROPERTIES OF AMORPHOUS ALLOYS

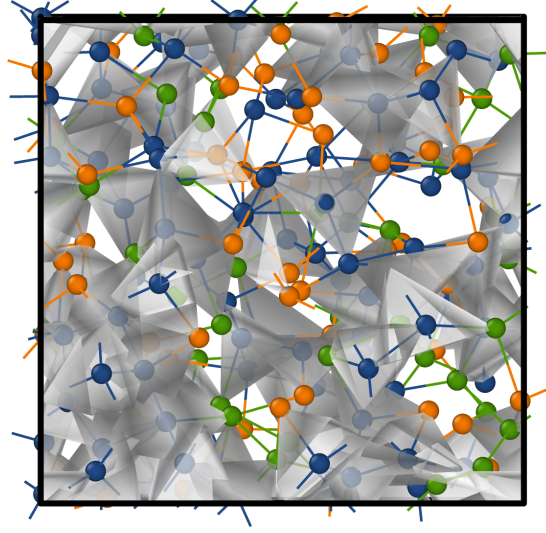


FIG. S2: Snapshot of a-GST523 at 300 K highlighting the presence of Ge in tetrahedral coordination. Ge atoms are shown in blue, Sb in green and Te in orange. Figure generated with the code Ovito<sup>6</sup>.

TABLE S2: Average coordination numbers for different pairs of atoms computed from the partial pair correlation functions in Ge-rich amorphous alloys (Fig. S3) and in a-GST225 (from Ref. 7).

	-	Total	With Ge	With Sb	With Te
a-GST523	Ge	4.06	1.82	0.83	1.41
	Sb	3.88	2.04	0.88	0.96
	Te	3.06	2.39	0.66	0.01
	-	Total	With Ge	With Sb	With Te
a-GST423	Ge	4.01	1.31	0.91	1.79
	Sb	3.80	1.82	1.00	0.98
	Te	3.04	2.39	0.65	0.00
	-	Total	With Ge	With Sb	With Te
a-GST323	Ge	4.00	0.82	0.96	2.22
	Sb	3.85	1.43	1.07	1.35
	Te	3.15	2.22	0.90	0.03
	-	Total	With Ge	With Sb	With Te
a-GST221	Ge	4.15	1.38	1.60	1.17
	Sb	3.58	1.60	1.52	0.46
	Te	3.18	2.29	0.89	0.00
	-	Total	With Ge	With Sb	With Te
a-GST225	Ge	3.96	0.29	0.36	3.31
	Sb	4.15	0.36	0.43	3.36
	Te	2.97	1.33	1.34	0.30

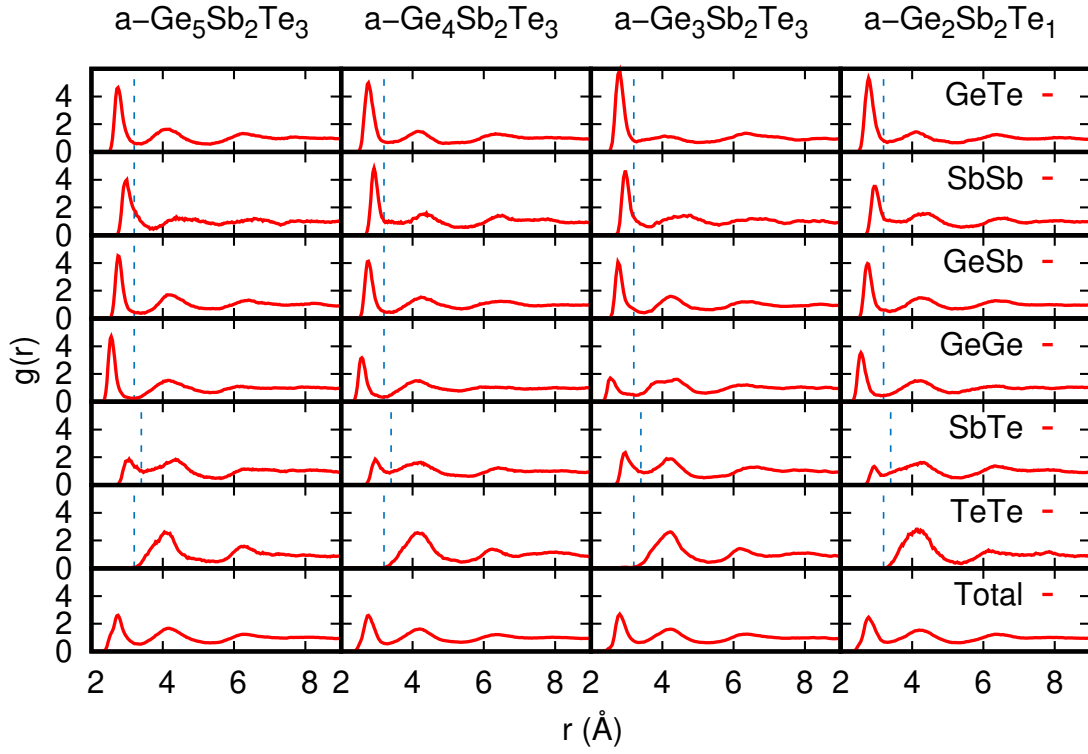


FIG. S3: Total and partial pair correlation functions of Ge-rich amorphous alloys at 300 K. The dashed lines indicate the bonding cutoff used to define the coordination numbers.

TABLE S3: Percentage fraction of the different types of bonds in Ge-rich amorphous alloys and in a-GST225 (from Ref. 7).

	-	Ge	Sb	Te
a-GST523	Ge	24.45	-	-
	Sb	22.20	4.81	-
	Te	37.92	10.53	0.1
	-	Ge	Sb	Te
a-GST423	Ge	15.98	-	-
	Sb	22.21	6.10	-
	Te	43.71	11.97	0.04
	-	Ge	Sb	Te
a-GST323	Ge	8.45	-	-
	Sb	19.67	7.35	-
	Te	45.67	18.52	0.34
	-	Ge	Sb	Te
a-GST221	Ge	14.78	-	-
	Sb	34.18	16.21	-
	Te	25.04	9.79	0.00
	-	Ge	Sb	Te
a-GST225	Ge	0.90	-	-
	Sb	7.90	2.80	-
	Te	43.40	45.10	4.10

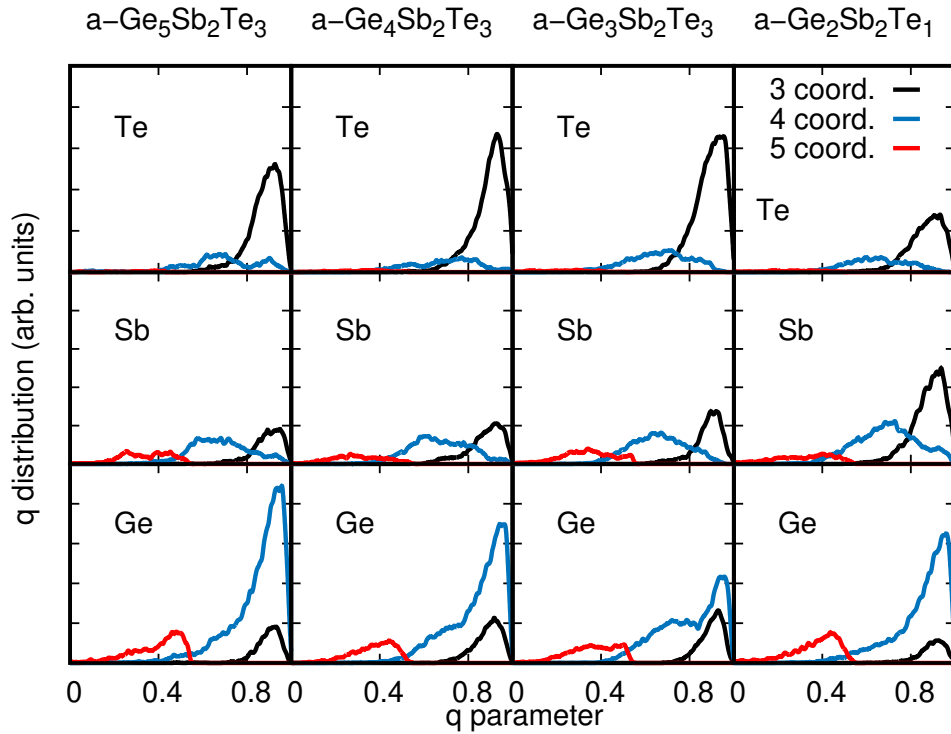


FIG. S4: Distribution of the local order parameter  $q$  resolved for atomic species and coordination number for Ge-rich amorphous alloys. To obtain a continuous distribution, the order parameter for individual atoms is broadened with a Gaussian function 0.005 wide.

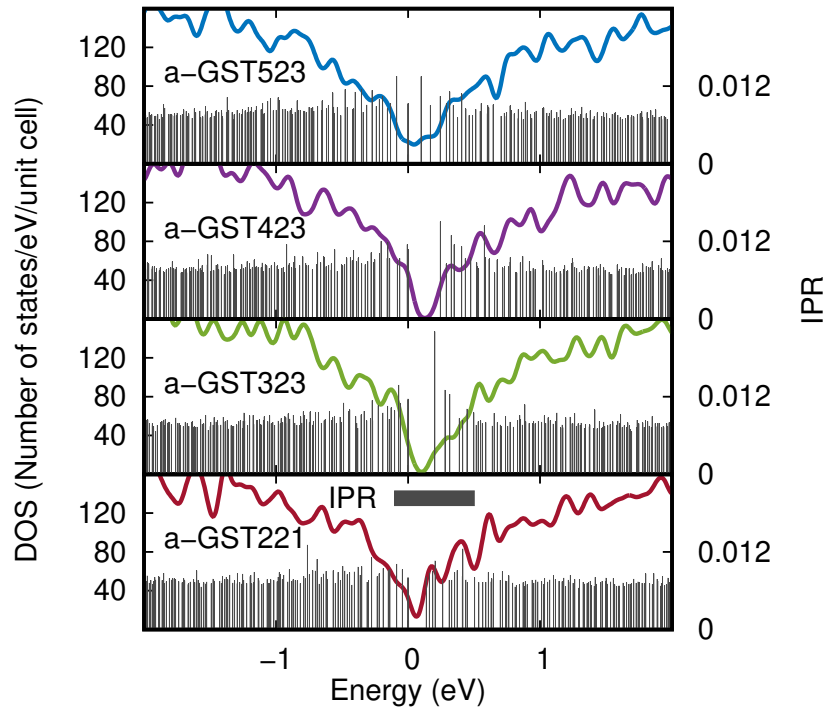


FIG. S5: Inverse Participation Ratio (IPR, gray spikes) superimposed to the electronic DOS of Ge-rich amorphous alloys. The DOS were computed with CP2k at the supercell  $\Gamma$  point only. The IPR for the  $i$ -th KS state is defined by  $\sum_j c_{ij}^4 / (\sum_j c_{ij}^2)^2$ , where  $j$  runs over the Gaussian-Type Orbitals (GTOs) of the basis set, while  $c_{ij}$  are the expansion coefficients of the  $i$ -th KS state in GTOs. The zero energy is the highest occupied KS state. The states close to the band edges are clearly more localized than the states deep in the valence and conduction bands.

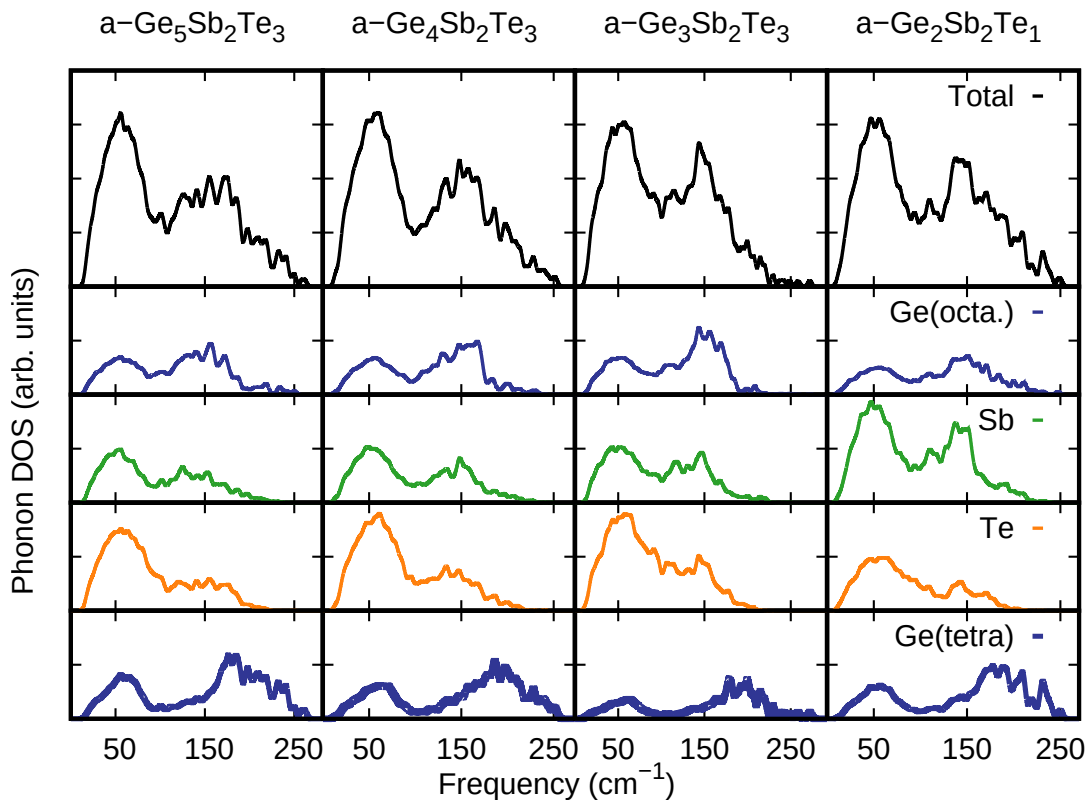


FIG. S6: The phonon DOS and the projection on different types of atoms for the Ge-rich amorphous alloys. The DOS is obtained from  $\Gamma$ -point phonon energies broadened by a Gaussian function with a variance of  $1.5 \text{ cm}^{-1}$ .

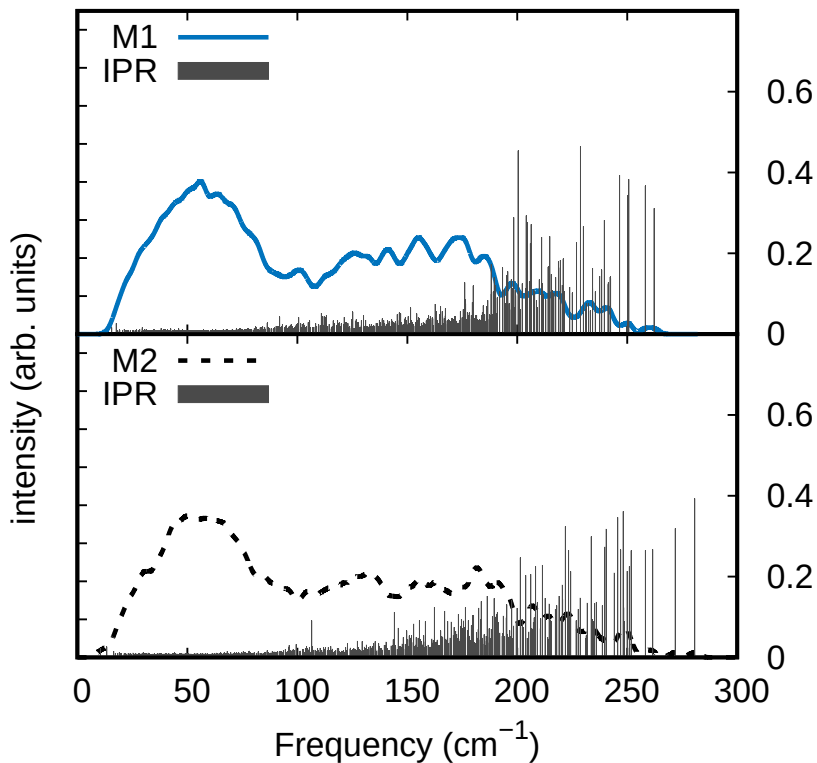


FIG. S7: Phonon DOS and IPR (gray spikes) of the two models of  $a\text{-GST523}$ .



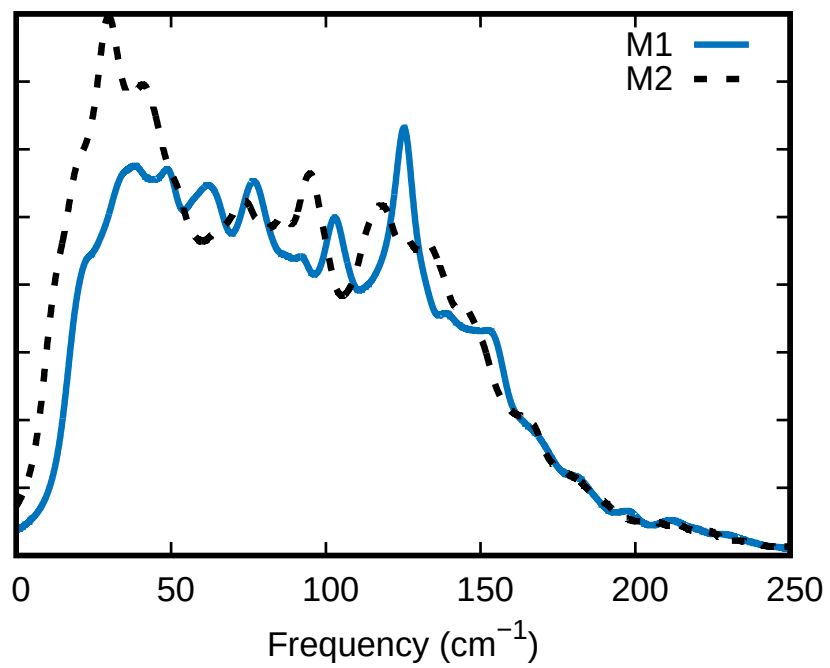


FIG. S8: Raman spectra of the two models of a-GST523 calculated with Lorentzian smearing of  $4 \text{ cm}^{-1}$ .

TABLE S4: Distribution of Ge, Sb and Te coordination environments for atoms with different coordination number in a-GST523. Environments in a fraction less than 1% are not reported.

Central atom	Coordination number				
	2	3	4	5	
Ge:		Ge <sub>2</sub> Sb 1.13%	Ge <sub>4</sub> 2.01%	Ge <sub>4</sub> Sb 1.55%	
		Ge <sub>2</sub> Te 1.18%	Ge <sub>3</sub> Sb 3.77%	Ge <sub>3</sub> SbTe 1.67%	
		GeTe <sub>2</sub> 4.34%	Ge <sub>2</sub> Sb <sub>2</sub> 7.64%	Ge <sub>2</sub> SbTe <sub>2</sub> 2.43%	
		Te <sub>3</sub> 1.26%	GeSb <sub>2</sub> Te 7.28%	GeSbTe <sub>3</sub> 1.86%	
			Sb <sub>2</sub> Te <sub>2</sub> 1.35%	Ge <sub>3</sub> Te <sub>2</sub> 1.67%	
			Ge <sub>2</sub> SbTe 14.3%	Ge <sub>2</sub> Te <sub>3</sub> 1.44%	
			GeSbTe <sub>2</sub> 11.5%		
			SbTe <sub>3</sub> 1.65%		
			Ge <sub>3</sub> Te 10.7%		
			Ge <sub>2</sub> Te <sub>2</sub> 10.4%		
			GeTe <sub>3</sub> 1.35%		
	Sb:		Ge <sub>3</sub> 3.88%	Ge <sub>4</sub> 6.27%	Ge <sub>2</sub> Sb <sub>2</sub> Te 2.18%
			Ge <sub>2</sub> Sb 12.1%	Ge <sub>3</sub> Sb 9.79%	Ge <sub>3</sub> SbTe 2.09%
		GeSb <sub>2</sub> 5.74%	Ge <sub>2</sub> Sb <sub>2</sub> 5.43%	Ge <sub>2</sub> SbTe <sub>2</sub> 5.75%	
		GeSbTe 4.81%	GeSb <sub>2</sub> Te 4.19%	Ge <sub>2</sub> Te <sub>3</sub> 4.09%	
		Ge <sub>2</sub> Te 1.99%	Ge <sub>2</sub> SbTe 6.75%	GeTe <sub>4</sub> 1.12%	
		GeTe <sub>2</sub> 1.25%	GeSbTe <sub>2</sub> 2.17%		
			Ge <sub>3</sub> Te 1.90%		
			Ge <sub>2</sub> Te <sub>2</sub> 7.59%		
Te:	Ge <sub>2</sub> 11.9%	Ge <sub>3</sub> 28.2%	Ge <sub>4</sub> 7.55%		
	GeSb 3.60%	Ge <sub>2</sub> Sb 29.4%	Ge <sub>3</sub> Sb 5.30%		
		GeSb <sub>2</sub> 4.49%	Ge <sub>2</sub> Sb <sub>2</sub> 4.76%		
			GeSb <sub>3</sub> 2.11%		

TABLE S5: Distribution of Ge, Sb and Te coordination environments for atoms with different coordination number in a-GST423. Environments in a fraction less than 1% are not reported.

Central atom	Coordination number			
	2	3	4	5
Ge:		GeTe <sub>2</sub> 3.42%	Ge <sub>4</sub> 1.39%	Ge <sub>3</sub> SbTe 1.97%
		SbTe <sub>2</sub> 3.09%	Ge <sub>3</sub> Sb 3.19%	Ge <sub>2</sub> SbTe <sub>2</sub> 2.55%
		Te <sub>3</sub> 5.30%	Ge <sub>3</sub> Te 4.43%	GeSb <sub>2</sub> Te <sub>2</sub> 1.00%
			Ge <sub>2</sub> Sb <sub>2</sub> 2.49%	GeSbTe <sub>3</sub> 1.10%
			Ge <sub>2</sub> SbTe 10.17%	GeTe <sub>4</sub> 1.20%
			Ge <sub>2</sub> Te <sub>2</sub> 8.69%	
			GeSb <sub>2</sub> Te 10.05%	
			GeSbTe <sub>2</sub> 7.84%	
			GeTe <sub>3</sub> 7.37%	
			Sb <sub>4</sub> 1.01%	
			Sb <sub>3</sub> Te 2.06%	
			Sb <sub>2</sub> Te <sub>2</sub> 3.23%	
			SbTe <sub>3</sub> 4.23%	
			Te <sub>4</sub> 3.67%	
	Sb:		Ge <sub>3</sub> 1.35%	Ge <sub>4</sub> 2.70%
		Ge <sub>2</sub> Sb 12.90%	Ge <sub>3</sub> Sb 12.66%	Ge <sub>3</sub> Te <sub>2</sub> 1.31%
		Ge <sub>2</sub> Te 3.82%	Ge <sub>3</sub> Te 4.45%	Ge <sub>2</sub> SbTe <sub>2</sub> 1.61%
		GeSb <sub>2</sub> 6.21%	Ge <sub>2</sub> Sb <sub>2</sub> 4.01%	Ge <sub>2</sub> Te <sub>3</sub> 1.27%
		GeSbTe 2.80%	Ge <sub>2</sub> SbTe 3.81%	GeSb <sub>2</sub> Te <sub>2</sub> 2.59%
		GeTe <sub>2</sub> 1.25%	Ge <sub>2</sub> Te <sub>2</sub> 8.28%	SbTe <sub>4</sub> 1.44%
		Sb <sub>2</sub> Te 3.60%	GeSb <sub>2</sub> Te 4.02%	
		SbTe <sub>2</sub> 1.82%	GeSbTe <sub>2</sub> 3.55%	
			GeTe <sub>3</sub> 1.22%	
			Sb <sub>3</sub> Te 1.43%	
Te:	Ge <sub>2</sub> 9.262%	Ge <sub>3</sub> 34.32%	Ge <sub>4</sub> 6.11%	
	GeSb 5.998%	Ge <sub>2</sub> Sb 23.98%	Ge <sub>3</sub> Sb 4.74%	
		GeSb <sub>2</sub> 7.355%	Ge <sub>2</sub> Sb <sub>2</sub> 4.02%	
			GeSb <sub>3</sub> 1.51%	

TABLE S6: Distribution of Ge, Sb and Te coordination environments for atoms with different coordination number in a-GST323. Environments in a fraction less than 1% are not reported.

Central atom	Coordination number				
	2	3	4	5	
Ge:		GeSbTe 1.88	Ge <sub>3</sub> Sb 2.46	Ge <sub>3</sub> SbTe 1.55	
		GeTe <sub>2</sub> 2.41	Ge <sub>2</sub> SbTe 8.86	Ge <sub>2</sub> SbTe <sub>2</sub> 2.18	
		SbTe <sub>2</sub> 4.79	Ge <sub>2</sub> Te <sub>2</sub> 3.72	Ge <sub>2</sub> Te <sub>3</sub> 2.28	
		Te <sub>3</sub> 8.52	GeSb <sub>2</sub> Te 4.57	GeSbTe <sub>3</sub> 2.00	
			GeSbTe <sub>2</sub> 9.53	GeTe <sub>4</sub> 2.31	
			GeTe <sub>3</sub> 9.79	SbTe <sub>4</sub> 2.50	
			Sb <sub>3</sub> Te 7.08		
			Sb <sub>2</sub> Te <sub>2</sub> 6.86		
			SbTe <sub>3</sub> 7.60		
			Te <sub>4</sub> 3.50		
	Sb:		Ge <sub>2</sub> Sb 4.22	Ge <sub>4</sub> 3.12	Ge <sub>3</sub> Te <sub>2</sub> 2.00
			Ge <sub>2</sub> Te 6.38	Ge <sub>3</sub> Sb 3.52	Ge <sub>2</sub> Sb <sub>2</sub> Te 1.11
			GeSb <sub>2</sub> 3.71	Ge <sub>3</sub> Te 5.34	Ge <sub>2</sub> SbTe <sub>2</sub> 1.95
		GeSbTe 6.25	Ge <sub>2</sub> Sb <sub>2</sub> 5.06	GeSb <sub>2</sub> Te <sub>2</sub> 2.36	
		GeTe <sub>2</sub> 2.43	Ge <sub>2</sub> SbTe 1.83	GeSbTe <sub>3</sub> 1.90	
		Sb <sub>3</sub> 3.75	Ge <sub>2</sub> Te <sub>2</sub> 3.17	GeTe <sub>4</sub> 1.96	
		Sb <sub>2</sub> Te 1.83	GeSb <sub>3</sub> 1.19	SbTe <sub>4</sub> 2.40	
		SbTe <sub>2</sub> 4.27	GeSb <sub>2</sub> Te 2.00		
			GeSbTe <sub>2</sub> 10.3		
			GeTe <sub>3</sub> 4.00		
			Sb <sub>2</sub> Te <sub>2</sub> 2.43		
		SbTe <sub>3</sub> 2.50			
Te:	Ge <sub>2</sub> 5.81	Ge <sub>3</sub> 27.9	Ge <sub>4</sub> 4.94		
	GeSb 3.23	Ge <sub>2</sub> Sb 26.7	Ge <sub>3</sub> Sb 7.82		
		GeSb <sub>2</sub> 9.11	Ge <sub>2</sub> Sb <sub>2</sub> 3.24		
		Sb <sub>3</sub> 2.52	GeSb <sub>3</sub> 3.96		

TABLE S7: Distribution of Ge, Sb and Te coordination environments for atoms with different coordination number in a-GST221. Environments in a fraction less than 1% are not reported.

Central atom	Coordination number			
	2	3	4	5
Ge:		GeSbTe 1.05%	Ge <sub>3</sub> Sb 4.55%	Ge <sub>3</sub> Te <sub>2</sub> 1.40%
		GeTe <sub>2</sub> 1.96%	Ge <sub>2</sub> Sb <sub>2</sub> 12.81%	Ge <sub>2</sub> Sb <sub>3</sub> 1.13%
		SbTe <sub>2</sub> 1.50%	Ge <sub>2</sub> SbTe 5.47%	Ge <sub>2</sub> Sb <sub>2</sub> Te 2.82%
		Te <sub>3</sub> 1.86%	Ge <sub>2</sub> Te <sub>2</sub> 4.58%	Ge <sub>2</sub> SbTe <sub>2</sub> 2.57%
			GeSb <sub>3</sub> 7.69%	GeSb <sub>3</sub> Te 1.57%
			GeSb <sub>2</sub> Te 15.2%	GeSb <sub>2</sub> Te <sub>2</sub> 1.61%
			GeSbTe <sub>2</sub> 6.54%	GeSbTe <sub>3</sub> 1.41%
			GeTe <sub>3</sub> 4.28%	
			Sb <sub>4</sub> 2.25%	
			Sb <sub>3</sub> Te 1.86%	
			Sb <sub>2</sub> Te <sub>2</sub> 2.34%	
			SbTe <sub>3</sub> 1.67%	
	Sb:		Ge <sub>3</sub> 5.28%	Ge <sub>4</sub> 2.59%
		Ge <sub>2</sub> Sb 9.94%	Ge <sub>3</sub> Sb 4.97%	Ge <sub>3</sub> Sb <sub>2</sub> 1.21%
		Ge <sub>2</sub> Te 3.19%	Ge <sub>3</sub> Te 2.68%	
		GeSb <sub>2</sub> 13.41%	Ge <sub>2</sub> Sb <sub>2</sub> 10.37%	
		GeSbTe 6.45%	Ge <sub>2</sub> SbTe 2.74%	
		Sb <sub>3</sub> 4.01%	GeSb <sub>3</sub> 5.59%	
		Sb <sub>2</sub> Te 2.75%	GeSb <sub>2</sub> Te 6.05%	
		GeSbTe <sub>2</sub> 2.06%		
		Sb <sub>3</sub> Te 1.67%		
Te:	Ge <sub>2</sub> 5.88%	Ge <sub>3</sub> 21.34%	Ge <sub>4</sub> 5.56%	Ge <sub>3</sub> Sb <sub>2</sub> 1.27%
	GeSb 6.06%	Ge <sub>2</sub> Sb 28.66%	Ge <sub>3</sub> Sb 11.67	
	Sb <sub>2</sub> 1.24%	GeSb <sub>2</sub> 7.31%	Ge <sub>2</sub> Sb <sub>2</sub> 7.52%	
			GeSb <sub>3</sub> 1.33%	

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- <sup>1</sup> A. Zunger, S.-H. Wei, L.G. Ferreira and J. E. Bernard, Special quasirandom structures, *Phys. Rev. Lett.* **65**, 353 (1990).
  - <sup>2</sup> C. S. Barrett, P. Cucka, and K. Haefner. The crystal structure of antimony at 4.2, 78 and 298 K, *Acta Crystallogr.* **16**, 451 (1963).
  - <sup>3</sup> P. Giannozzi et al., QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials, *J. Phys.: Condens. Matter* **21**, 395502 (2009); [www.quantum-espresso.org](http://www.quantum-espresso.org).
  - <sup>4</sup> J. Goldak, C. S. Barrett, D. Innes, and W. Youdelis, Structure of alpha GeTe, *J. Chem. Phys.* **44**, 3323 (1966).
  - <sup>5</sup> T.L. Anderson and H.B. Krause, Refinement of the  $\text{Sb}_2\text{Te}_3$  and  $\text{Sb}_2\text{Te}_2\text{Se}$  structures and their relationship to nonstoichiometric  $\text{Sb}_2\text{Te}_{3-y}\text{Se}_y$  compounds, *Acta Crystallogr. B* **30**, 1307 (1974).
  - <sup>6</sup> A. Stukowski, Visualization and analysis of atomistic simulation data with OVITO - the Open Visualization Tool, *Modelling Simul. Mater. Sci. Eng.* **18**, 015012 (2010).
  - <sup>7</sup> S. Gabardi, S. Caravati, M. Bernasconi, and M. Parrinello, Density functional simulations of Sb-rich GeSbTe phase change alloys, *J. Phys.: Condens. Matter* **24**, 385803 (2012).