Supplementary information for:

Structure of amorphous $Ge_8Sb_2Te_{11}$: GeTe-Sb_2Te_3 alloys and optical storage

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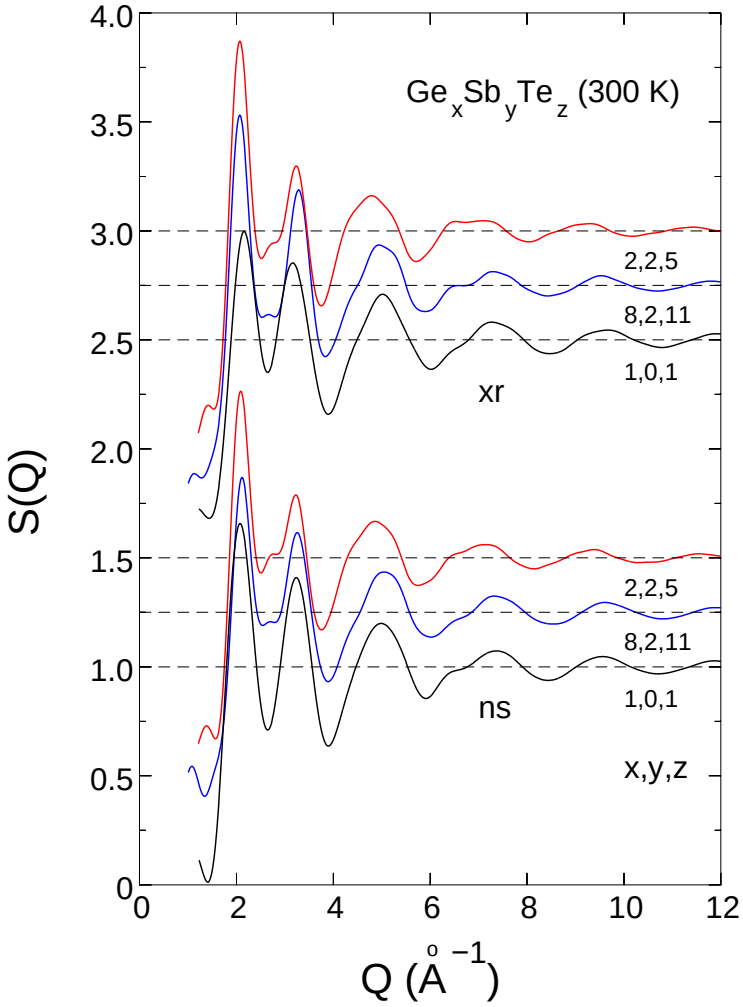

FIG. 1: [Supplementary Figure 1] Simulated x-ray (xr) and neutron scattering (ns) structure factors of $Ge_xSb_yTe_z$ alloys at 300 K. The curves have been displaced vertically for clarity.

The calculated structure factors $S(Q)$ of amorphous GST-225, GST-8,2,11, and GeTe are compared in Supplementary Figure 1. The $S(Q)$ are calculated as Fourier transforms of pair distribution functions (PDF) at 300 K using appropriate weights (x-ray, neutron scattering) for each elements.[1] The simulations for a-GST-225 and a-GeTe[1,2] were performed with the PBE approximation for the exchange-correlation energy functional.[3] The use of the PBEsol approximation in the present work leads to systematically shorter (1-2 %) bonds.[4] The simulations used similar melt-quench processes lasting several hundred picoseconds, but the sample sizes differ: a-GeTe: 216 atoms, a-GST-225: 460 atoms, a-GST-8,2,11: 630 atoms.

The PDF between atoms and cavity centers in a-GST-8,2,11 are plotted in Supplementary Figure 2. The cavity centers have been determined by mapping the points that are farthest

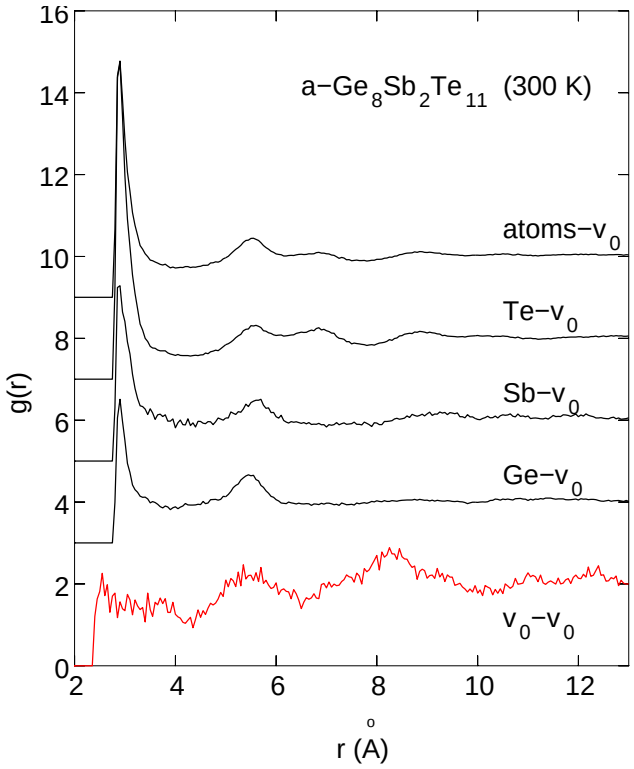

FIG. 2: [Supplementary Figure 2] Pair distribution functions between atoms and cavity centers (v_o) at 300 K. The scale of the lowest curve (vacancy-vacancy, red) has been doubled and the curves displaced vertically for clarity.

from any atom in a given cavity (i.e., the center of the largest test sphere). The cavity analysis is described in Ref. 1. Cavities reflect the underlying atomic structure, and their PDF show corresponding correlations. The most frequent atomic neighbor of cavities is Te (3.7 neighbors), and Sb is preferred over Ge, although the small Sb concentration (< 10 %) means that Ge occurs more often in the cavity neighborhood. The number of cavities fluctuates during the simulation at 300 K, and the 630-atom sample contains an average of 50. The distribution of cavities shows a monotonic decrease as a function of cavity volume, but there are several larger than 100 Å^3.

The projected density of states (DOS) of valence electrons is shown for c- and a-GST-8,2,11 in Supplementary Figure 3. The projection of Kohn-Sham wave functions has been performed with respect to the atomic orbitals according to atomic type (top panel) and angular momentum (middle panel). The two lowest (σ) bands can be divided into Te-5s, Sb-5s, and Ge-4s regions (see Fig. 8 in the text). The Sb-5s peak is considerably weaker

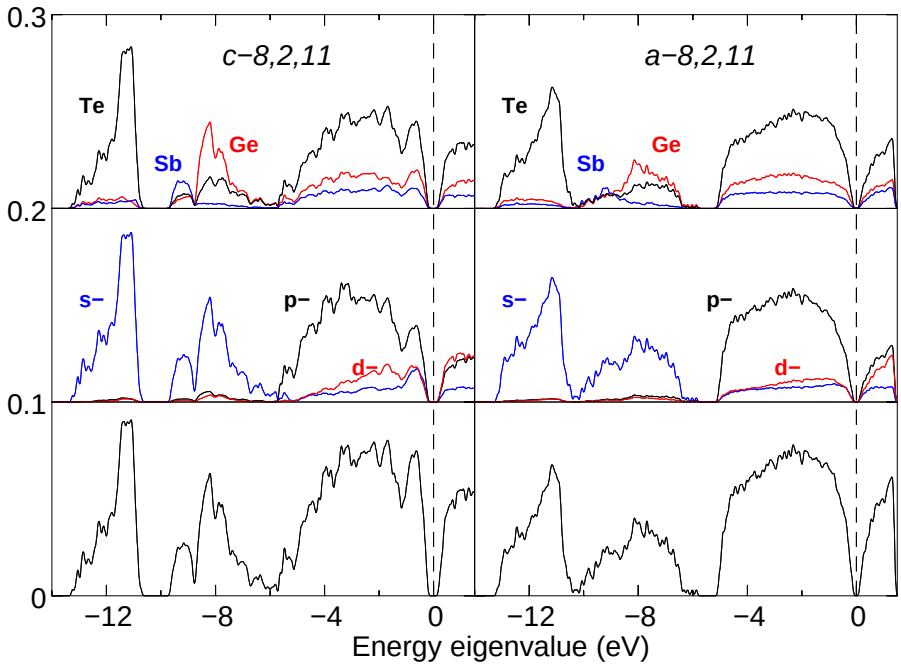

FIG. 3: [Supplementary Figure 3] Projected density of states for crystalline (left) and amorphous (right) phases of GST-8,2,11. The top panels show projections with respect to atoms, the middle panels with respect to atomic orbitals, and the lowest panels the total DOS.

than that in GST-225,[1] and a band gap between the two σ bands at -10 eV in a-GST-8,2,11 is almost present. The π-band is dominated by the most abundant element (Te) and atomic p-character. The d-character originates from Sb and Te, and it increases as the energy increases through the Fermi energy. The d-character is negligible in GeTe.

The effective atomic charges in GST-8,2,11 have been calculated by integrating the electron density over the atomic Voronoi cells, with no volume (and charge) assigned to the cavities. A separate integration is performed inside the cavities, and the results for GST-8,2,11 are compared with our previous results for GST-225[1] in Supplementary Figure 4. Sb is the most cationic of the three elements. The charge in the cavities originates mainly from the surrounding Te atoms and their lone pair orbitals. Ge is the smallest of the three elements and its Voronoi volume and the charge inside may be overestimated. However, the projected

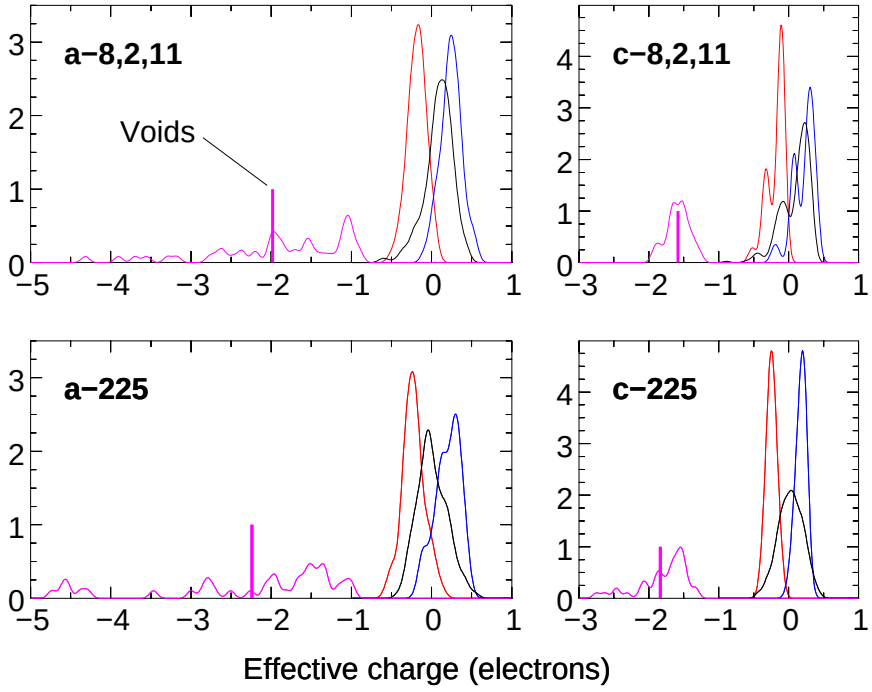

FIG. 4: [Supplementary Figure 4] Effective charge distributions in GST-8,2,11 and GST-225 (Ge: red, Sb: Blue, Te: black, and cavities: magenta). The charges have been calculated with the Voronoi prescription. The atomic charges do not include any contribution from the cavities. The vertical bar marks the average charge enclosed by the cavities.

Mulliken and Löwdin charges also display negative values for Ge.

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