ADVANCED FUNCTIONAL MATERIALS

Supporting Information

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Atomistic Study of the Configurational Entropy and the Fragility of Supercooled Liquid GeTe

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Supporting Information

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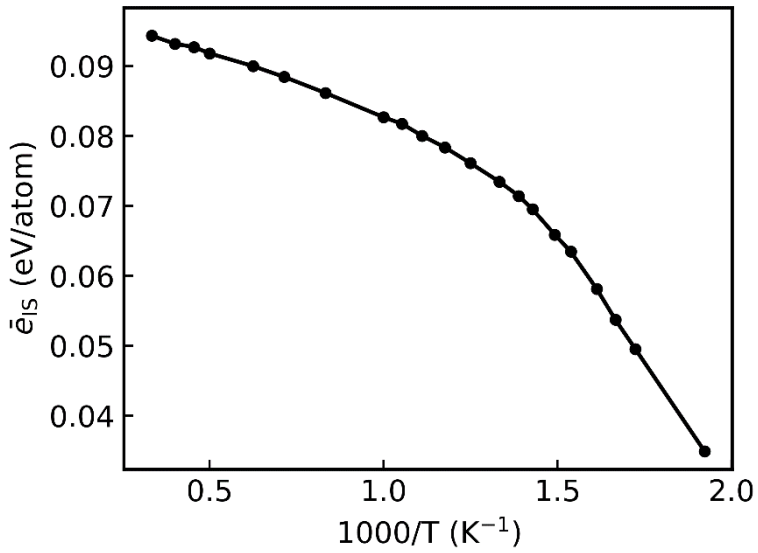

Figure S1. Average inherent structure energy $\bar{e}_{IS}$ as a function of $1/T$. The temperature is scaled by a factor of 1000.

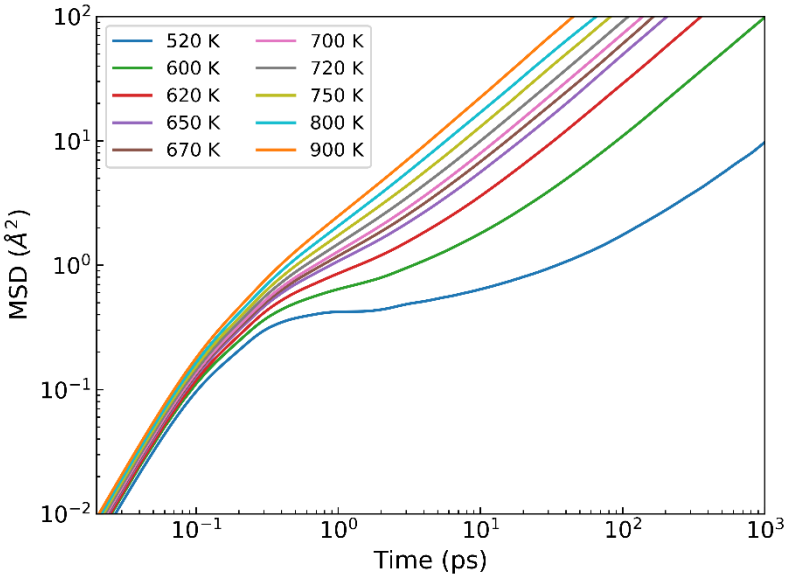

Figure S2. The mean square displacement (MSD) of supercooled liquid GeTe at temperatures from 520 K to 900 K. At each temperature the MSD is calculated from $\frac{1}{N}\sum_{i=1}^{N}\langle(\boldsymbol{r}_i(t)-\boldsymbol{r}_i(t_0))^2\rangle$, where N denotes the number of atoms. The curves at temperatures below 650 K show plateaus due to the caging effect. All the curves raise again after the plateau, which is indicative of α relaxation.

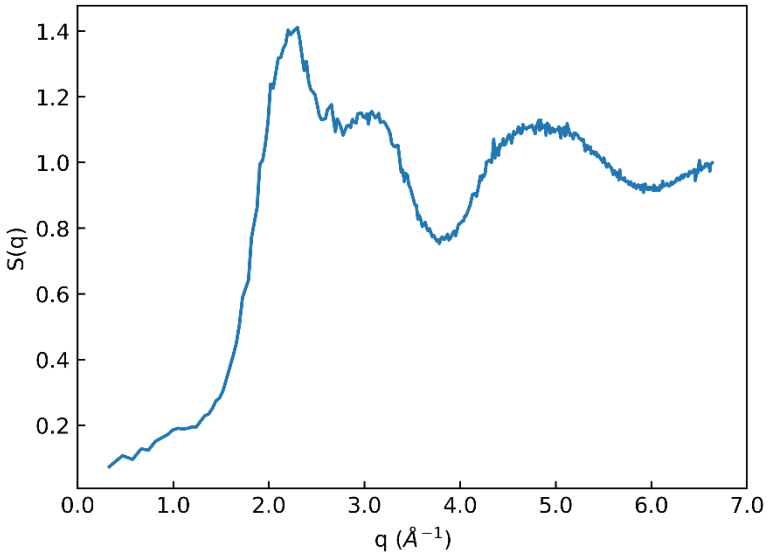

Figure S3. Static structure factor $S(q)$ of liquid GeTe at 1000 K. The first peak is centered at $q_0 = 2.2$ Å^{-1}. The structure factor was calculated as $S(q) = N^{-1}\langle\rho(\mathbf{q})\rho(-\mathbf{q})\rangle$, where $\rho(\mathbf{q}) = \sum_{i=1}^{N}\exp(-i\mathbf{q}\cdot\mathbf{r}_i)$ is the spatial Fourier transform of the number density.

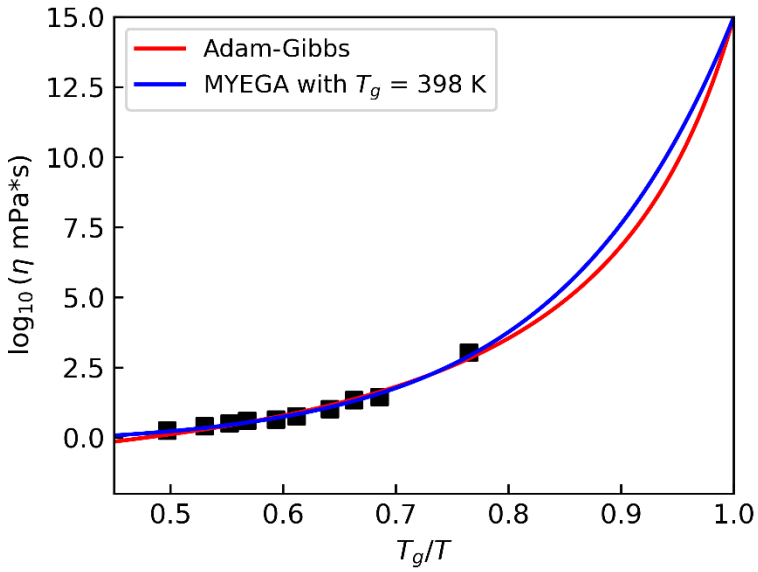

Figure S4. Comparison between the viscosity curve obtained from the AG relation and the explicit calculation of the configurational entropy (red curve corresponding to Fig. 4(a) in the main text) and the curve obtained from the MYEGA equation (blue curve). The MYEGA equation is based on the Adam-Gibbs relation and the constraint theory for the modeling of the configurational entropy [1]. It is widely used to fit the temperature dependence of the viscosity from both experimental [2,3] and computational [4] data. The MYEGA equation for the viscosity η reads:

$$\log_{10}\eta(T) = \log_{10}\eta_0 + (15 - \log_{10}\eta_0)\frac{T_g}{T}\exp\left[\left(\frac{m}{15 - \log_{10}\eta_0} - 1\right)\left(\frac{T_g}{T} - 1\right)\right],$$

where T_g is an input parameter (which we set to T_g = 398 K) and both η_0 and the fragility index m are fitting parameters. The fit of the viscosity values calculated with the Green-Kubo formula (black squares in the figure) with the MYEGA equation yields the blue curve and a fragility index $m = 100$ ($\log_{10}\eta_0 = -0.24$).

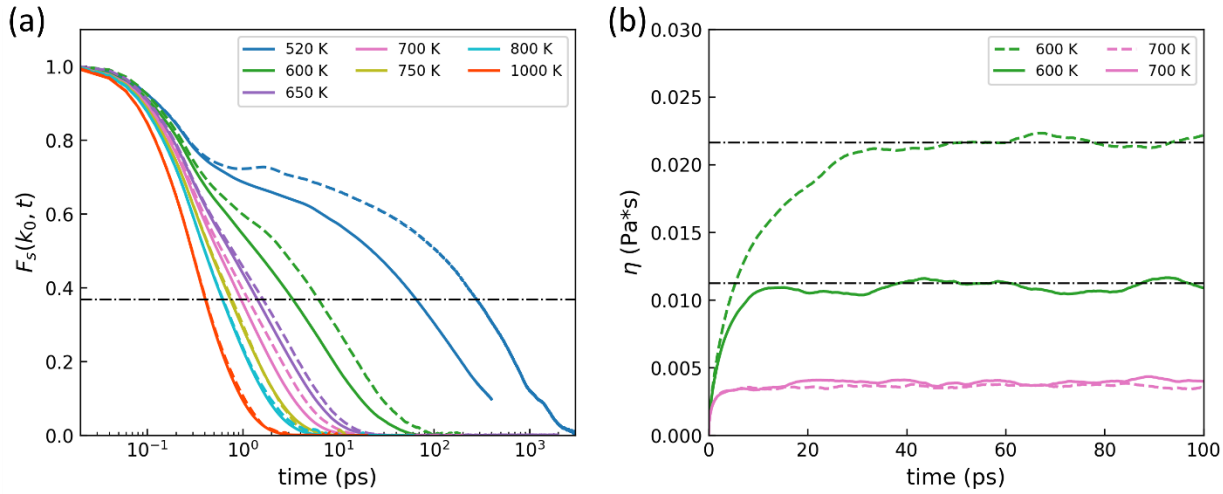

Figure S5. (a) The incoherent intermediate scattering function of GeTe from 520 K to 1000 K. The x axis has logarithmic scale. Dashed and continuous lines correspond to 216-atom and 4096-atom models, respectively. At low temperature, the large models display shorter relaxation times. (b) Convergence plots of the viscosity at 600 K and 700 K obtained from the Green-Kubo formula (formula (11) in the main text). Dashed and continuous lines correspond to 216-atom and 4096-atom models. The black dashed-dotted lines correspond to the converged values of the viscosity at 600 K. At this temperature, there are significant finite-size effects.

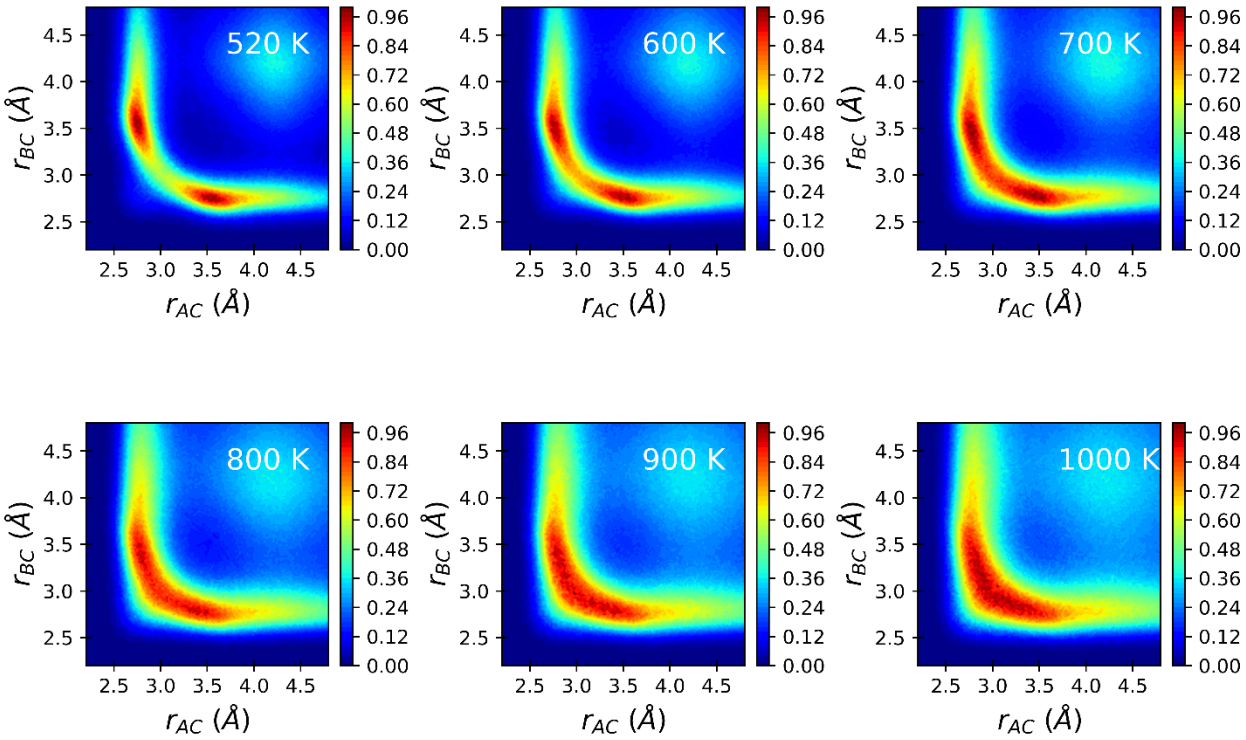

Figure S6. Angular-limited three-body correlation function (ALTBC) of liquid GeTe as a function of temperature. The ALTBC provides information on the two-dimensional correlation of the interatomic distances for triplets of atoms that are almost aligned. We considered inter-bond angles in the range between 155° and 180°. Two separate maxima symmetric with respect to the diagonal can be seen up to 800 K, indicating Peierls-like distortion. At 900 K and above, the two peaks appear to merge, yielding a single, stretched peak centered on the diagonal.

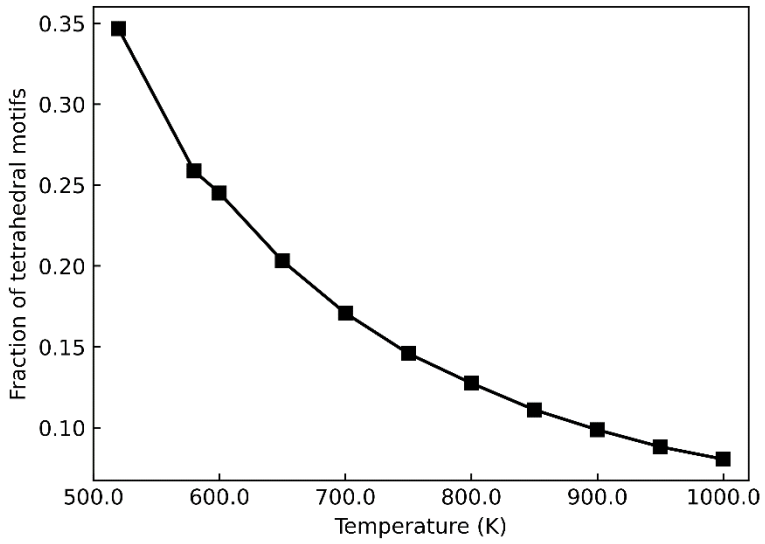

Figure S7. Fraction of Ge atoms with tetrahedral coordination in liquid GeTe as a function of temperature.

References

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