Viscosity, breakdown of Stokes-Einstein relation and dynamical heterogeneity in supercooled liquid $Ge_2Sb_2Te_5$ from simulations with a neural network potential

Supplementary Material

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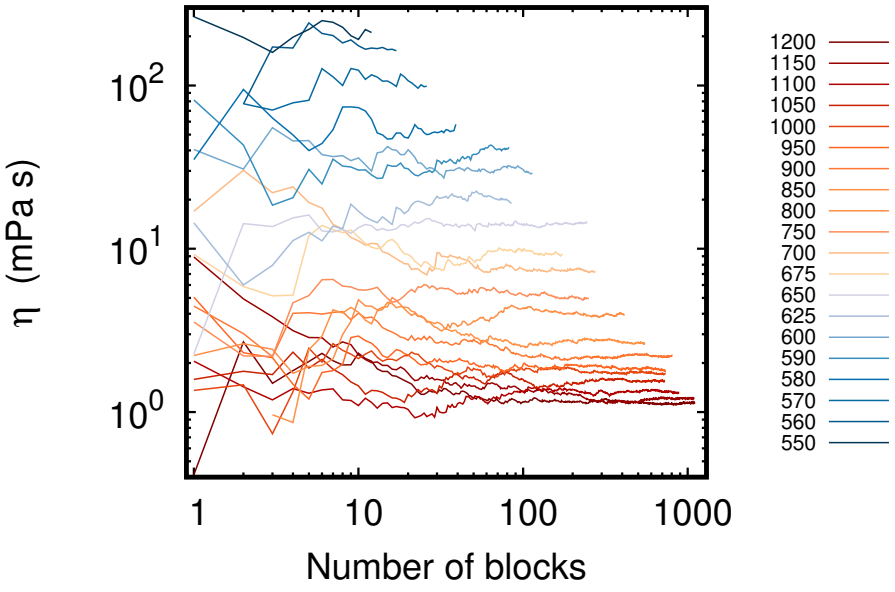

Figure S1. Convergence of the viscosity $\eta(t_{max})$ at time t_{max} as a function of the number of blocks ($2t_{max}$ long) used in the block average (see Section II in the article).

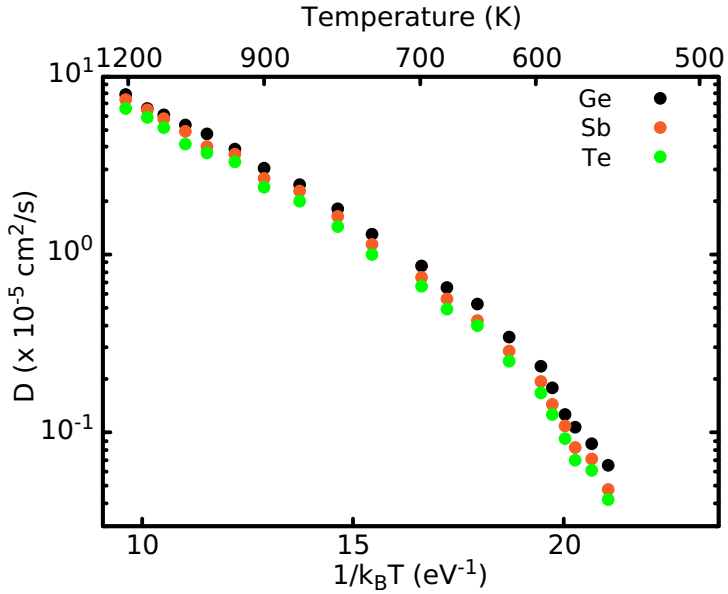

Figure S2. Diffusion coefficient as a function of temperature resolved for the different species.

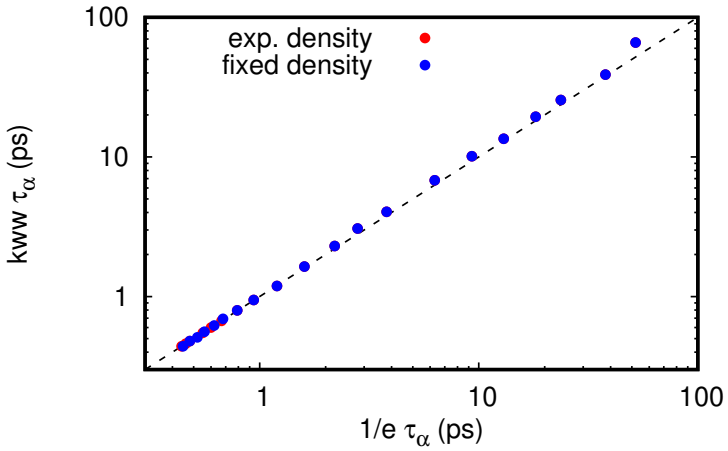

Figure S3. Comparison of τ_α obtained from the KWW fit at long times and from the condition $F_s(q_o,t)= 1/e$. The data refer to different temperatures.

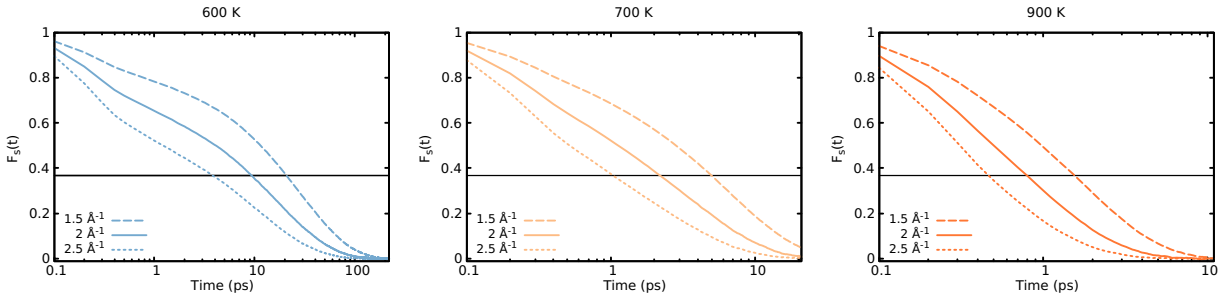

Figure S4. Incoherent intermediate scattering function $F_s(q,t)$ for three different values of q and at the three different temperatures.

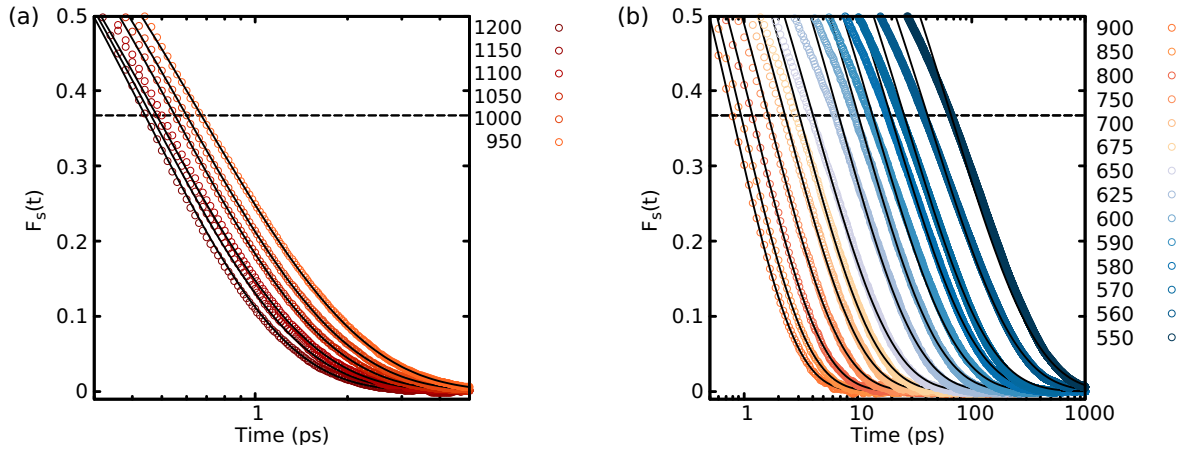

Figure S5. KWW fit (continuous lines) of the ISF (open dots, see Figure 2 in the article) for temperatures (a) above T_m and (b) below T_m.

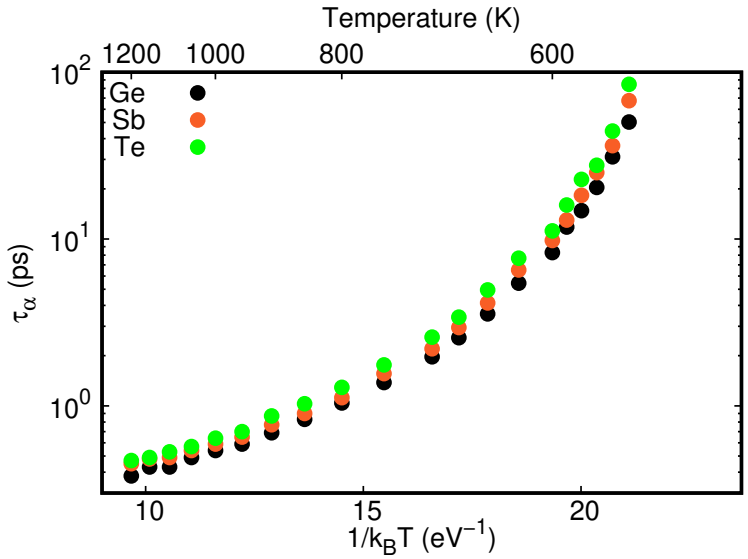

Figure S6. The relaxation time τ_α as a function of temperature resolved for the different species. For the data above T_m we used the values at the experimental density.

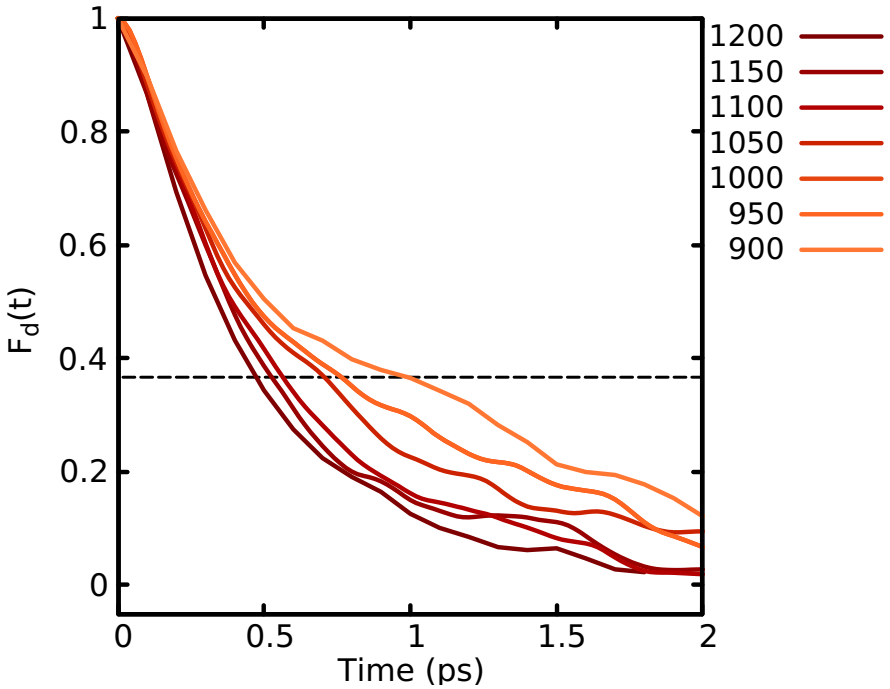

Figure S7. Coherent (distinct) intermediate scattering function $F_d(q_o,t)$ as a function of time at different temperatures above T_m from NVT simulations. We chose q_o = 2 Å^{-1} that corresponds to the main peak in the experimental static structure factor (see article). The horizontal line indicates the condition $F_d(q_o,t) = 1/e$.

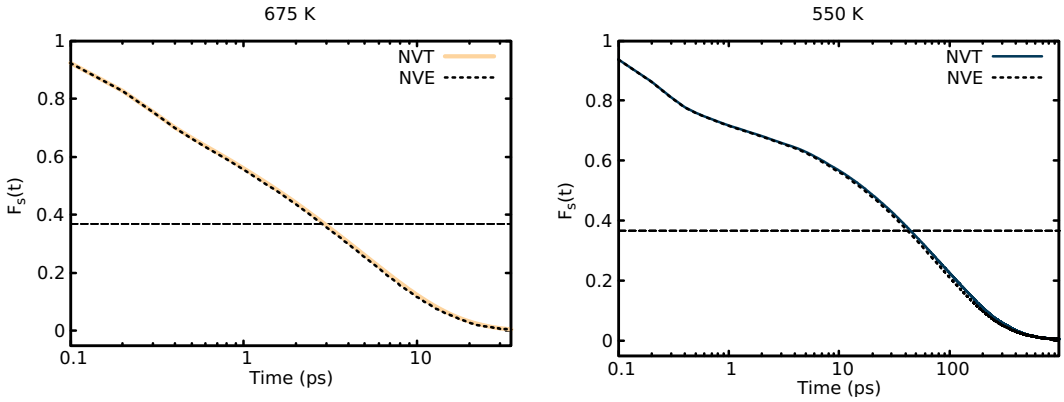

Figure S8. Incoherent intermediate scattering functions $F_s(q_o,t)$ obtained from NVT and NVE simulations, of the same length, at two different temperatures.

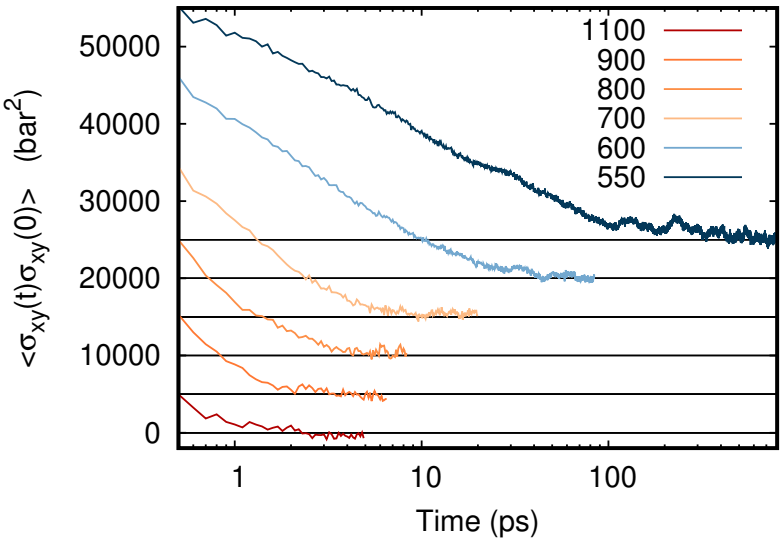

Figure S9. The self-correlation function $\langle\sigma_{xy}(t)\sigma_{xy}(0)\rangle$ entering in the Green-Kubo integral for the evaluation of the viscosity (see article) for a few representative temperatures. The curves are displaced vertically by 5000 bar^2 each, to improve the readability of the figure.

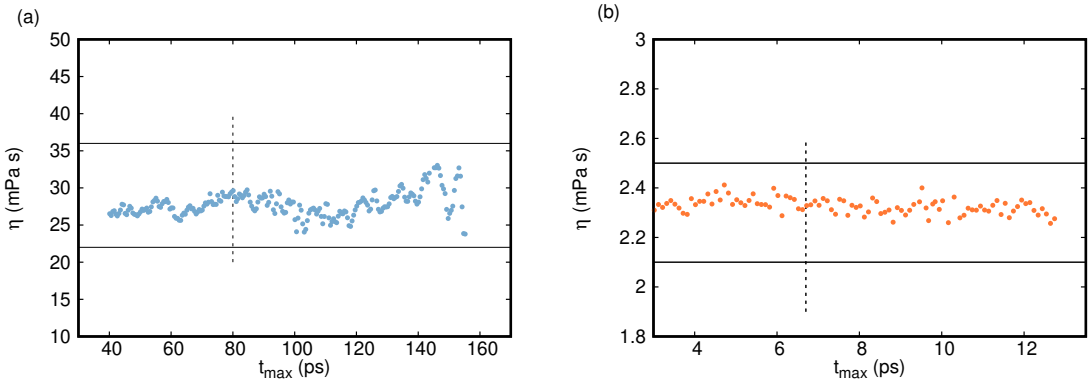

Figure S10. Dependence of our evaluation of η on the choice of the maximum integration time t_{max} in the GK formula (Eq. (1) in the article) at low (600 K, left panel) and high (900 K, right panel) temperatures. The horizontal lines refer to the highest and lowest estimation of η given our error-bars, while the vertical dashed line corresponds to our choice of t_{max}.

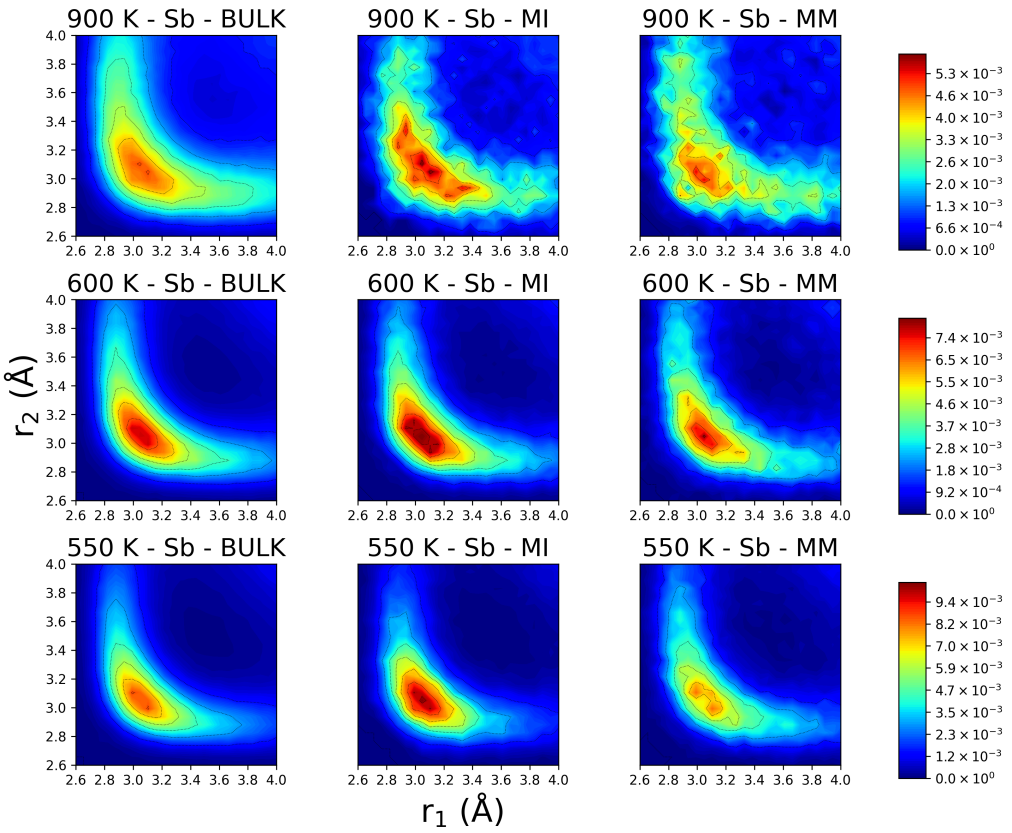

Figure S11. ALTBC function for Sb atoms at (top panels) 900 K (central panels) 600 K and (lower panels) 550 K for the bulk (left panels), the two largest MI clusters (central panels) and the two largest MM clusters (right panels).

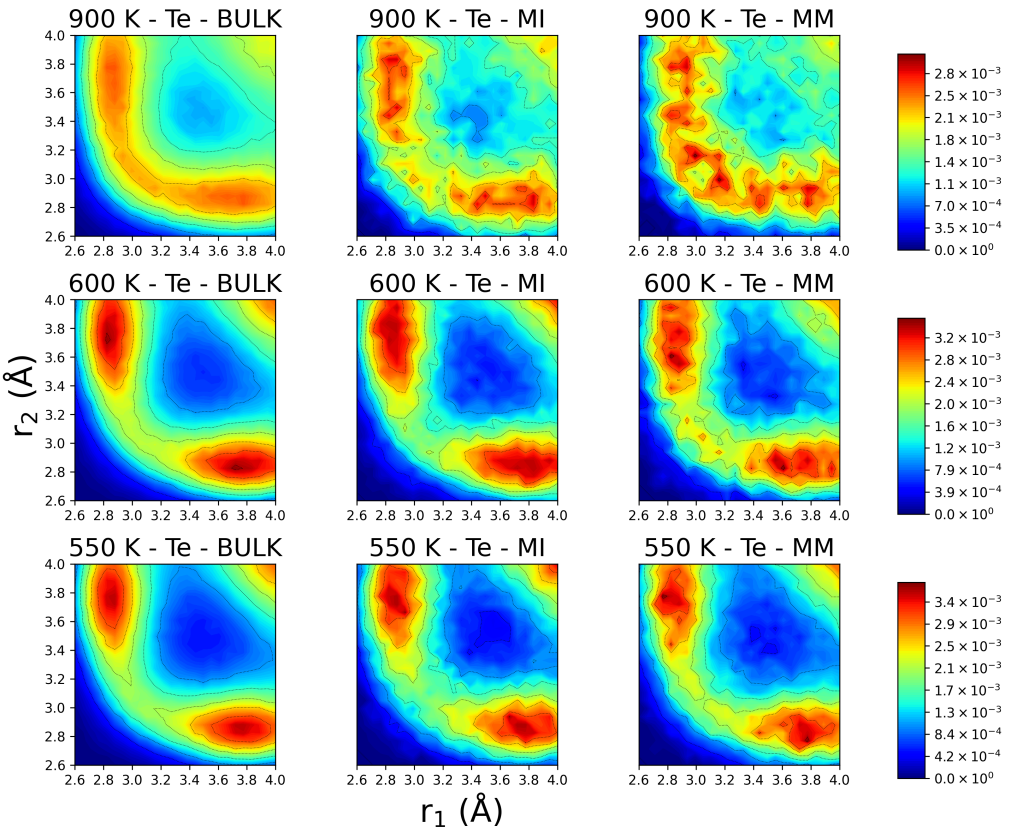

Figure S12. ALTBC function for Te atoms at (top panels) 900 K (central panels) 600 K and (lowe panels) 550 K for the bulk (left panels), the two largest MI clusters (central panels) and the two largest MM clusters (right panels).

Temperature (K)	simulation ensemble	density (atom/Å^3)	simulation time (ns)	t_{max} (ps)	η (mPa s)	D_{Ge} (10^{-5} cm^2/s)	D_{Sb} (10^{-5} cm^2/s)	D_{Te} (10^{-5} cm^2/s)
1200	NVT	2.993	6.6	3	η=1.15 $\pm$ 0.07 *			
1203	NVE	2.993	0.12			7.84	7.36	6.62
1200	NVT	3.075	6.6	3.2	η=1.20 $\pm$ 0.08 *			
1208	NVE	3.075	0.12			7.29	6.95	6.19
1150	NVT	3.009	8.8	4	η=1.20 $\pm$ 0.06 *			
1150	NVE	3.009	0.12			6.64	6.45	5.83
1150	NVT	3.075	8.8	4.2	η=1.31 $\pm$ 0.08 *			
1149	NVE	3.075	0.12			6.44	6.11	5.42
1100	NVT	3.023	8.8	5	η=1.34 $\pm$ 0.08 *			
1107	NVE	3.023	0.2			6.12	5.76	5.12
1100	NVT	3.075	8.8	5	η=1.39 $\pm$ 0.07 *			
1104	NVE	3.075	0.2			5.72	5.38	4.81
1050	NVT	3.039	8.8	5	η=1.52 $\pm$ 0.06 *			
1043	NVE	3.039	0.2			5.32	4.93	4.17
1050	NVT	3.075	8.8	5.6	η=1.6 $\pm$ 0.1 *			
1053	NVE	3.075	0.2			5.28	4.53	4.09
1000	NVT	3.052	8.8	6	η=1.7 $\pm$ 0.1 *			
1000	NVE	3.052	0.2			4.72	3.98	3.74
1000	NVT	3.075	8.8	6.2	η=1.7 $\pm$ 0.08 *			
1006	NVE	3.075	0.2			4.40	3.88	3.63
950	NVT	3.063	8.8	6	η=1.8 $\pm$ 0.1 *			
957	NVE	3.063	0.2			3.87	3.64	3.32
950	NVT	3.075	8.8	6.4	η=1.9 $\pm$ 0.1 *			
952	NVE	3.075	0.2			3.73	3.64	3.13
900	NVT	3.075	8.8	6.6	η=2.3 $\pm$ 0.2 *			
902	NVE	3.075	0.2			3.05	2.66	2.40
850	NVT	3.075	8.8	8	η=2.7 $\pm$ 0.2 *			
846	NVE	3.075	0.6			2.35	2.29	1.82

Temperature (K)	simulation ensemble	density (atom/Å^3)	simulation time (ns)	t_{max} (ps)	η (mPa s)	D_{Ge} (10^{-5} cm^2/s)	D_{Sb} (10^{-5} cm^2/s)	D_{Te} (10^{-5} cm^2/s)
800	NVT	3.075	8.8	8.4	η=3.5 $\pm$ 0.5 *			
794	NVE	3.075	0.4			1.81	1.62	1.43
750	NVT	3.075	11	17.2	η=5.1 $\pm$ 0.7 *			
751	NVE	3.075	0.4			1.29	1.13	1.0
700	NVT	3.075	11	19.2	η=6.9 $\pm$ 0.9 *	0.87	0.75	0.66
698	NVE	3.075	0.4			0.866	0.749	0.659
675	NVT	3.075	11	24	η=9.5 $\pm$ 0.9 *			
674	NVE	3.075	0.4			0.652	0.558	0.495
650	NVT	3.075	13.2	28	η=13 $\pm$ 2 *			
647	NVE	3.075	0.6			0.525	0.426	0.398
625	NVT	3.075	13.2	76	η=13 $\pm$ 2 *			
621	NVE	3.075	0.55			0.341	0.285	0.248
600	NVT	3.075	13.2	84	η=29 $\pm$ 7 *			
597	NVE	3.075	0.6			0.237	0.194	0.168
590	NVT	3.075	17.6	140	η=39 $\pm$ 7 *			
588	NVE	3.075	1.0			0.178	0.143	0.127
580	NVT	3.075	17.6	180	η=57 $\pm$ 8 *			
580	NVE	3.075	2.0			0.126	0.109	0.092
570	NVT	3.075	18.0	199	η=90 $\pm$ 12 *			
573	NVE	3.075	2.0			0.108	0.083	0.070
560	NVT	3.075	18.0	300	η=153 $\pm$ 34 *			
562	NVE	3.075	2.0			0.086	0.071	0.061
550	NVT	3.075	18.0	800	η=218 $\pm$ 46 *			
551	NVE	3.075	2.0			0.065	0.048	0.042

Table SI. Synoptic table of the different simulations (see article) with temperature, simulation ensemble, density, simulation time, maximum time t_{max} used in the Green-Kubo integral for the viscosity, the resulting viscosity and specie resolved diffusion coefficients. The * indicates that η is obtained by block averaging over several simulations under the same conditions (see article), the corresponding simulation time is the overall time summed over the independent simulations.