Crystallization kinetics in Ge-rich Ge_xTe alloys from large scale simulations with a machine-learned interatomic potential

SUPPLEMENTARY MATERIAL

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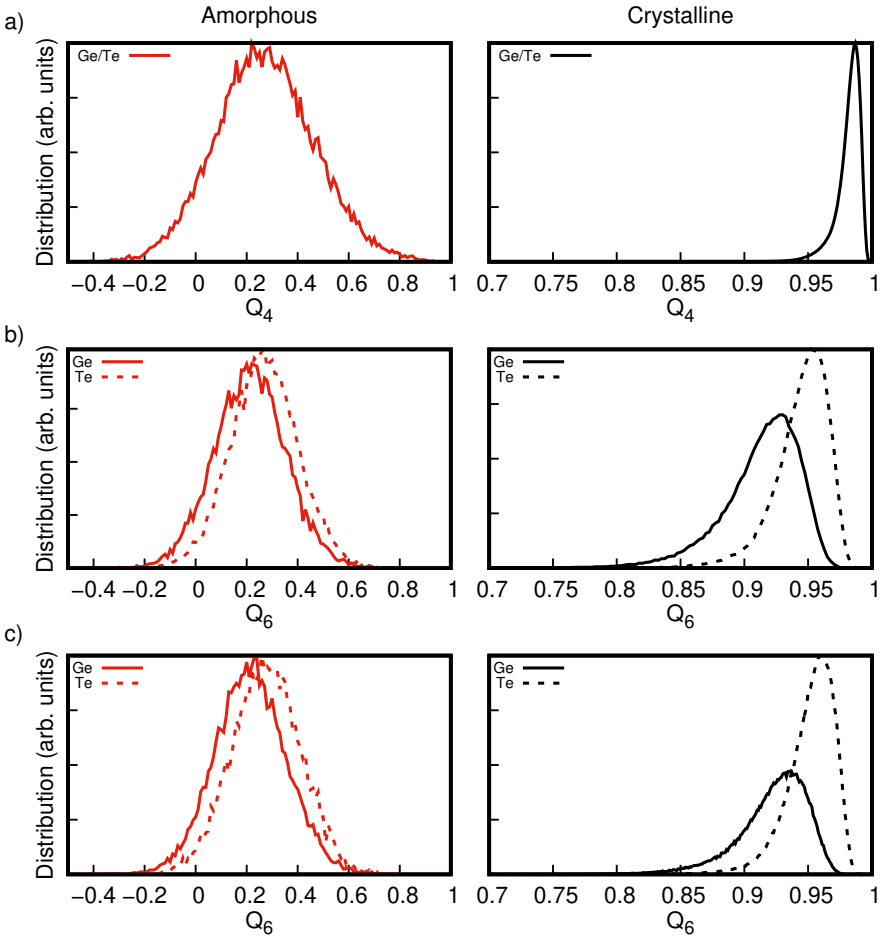

Figure S1: a) Distribution of the Q_4^{dot} parameter in amorphous and crystalline cubic (β-phase) GeTe at 600 K in 4096-atom models. An atom is considered crystalline for $Q_4^{dot} > 0.87$. b) Distribution of the Q_6^{dot} parameter for Ge (continuous line) and Te (dashed line) atoms in amorphous and crystalline cubic GeTe (β-phase) at 600 K. A Ge (Te) atom is considered crystalline for $Q_6^{dot} > 0.735$ (0.815). c) The same of panel b) at 500 K. A Ge (Te) atom is considered crystalline for $Q_6^{dot} > 0.75$ (0.83).

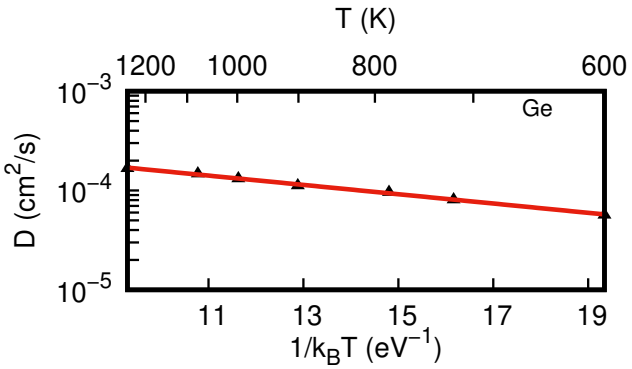

Figure S 2: Diffusion coefficient (D) as a function of temperature of pure Ge from NN simulations. The experimental value for D at 1273 K is 1.12 10^{-4} cm^2/s (S. Chathoth *et al.*, Appl. Phys. Lett. **94**, 221906 (2009)).

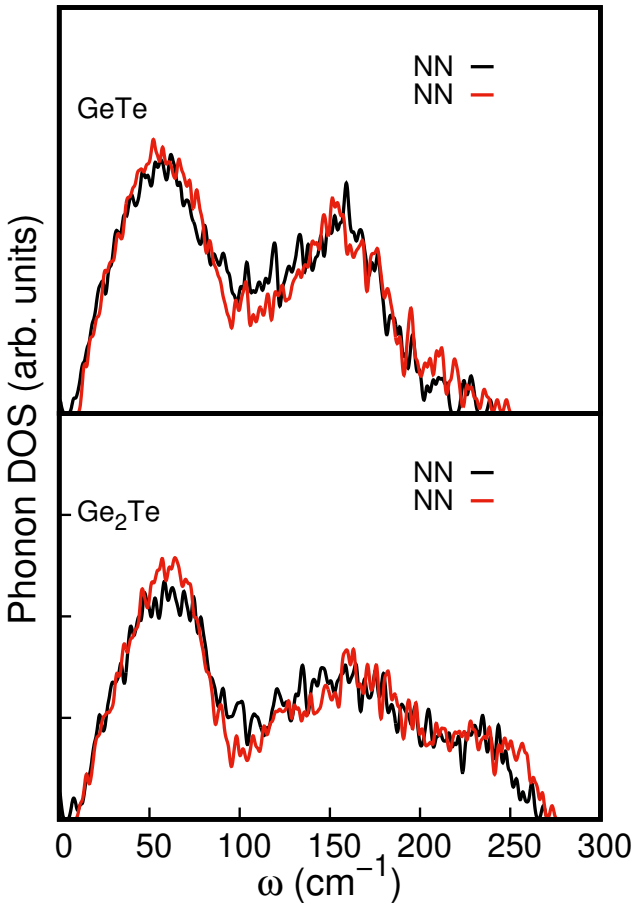

Figure S3: Phonon density of states (DOS) from NN and DFT simulations of the amorphous phase of GeTe and Ge_2Te in 300-atom cells. The DOS is obtained from Γ-point phonons whose energies are broadened with a Gaussian with variance 1 cm^{-1}. The dynamical matrix is built from finite difference of forces under atomic displacements of 0.005 Å.

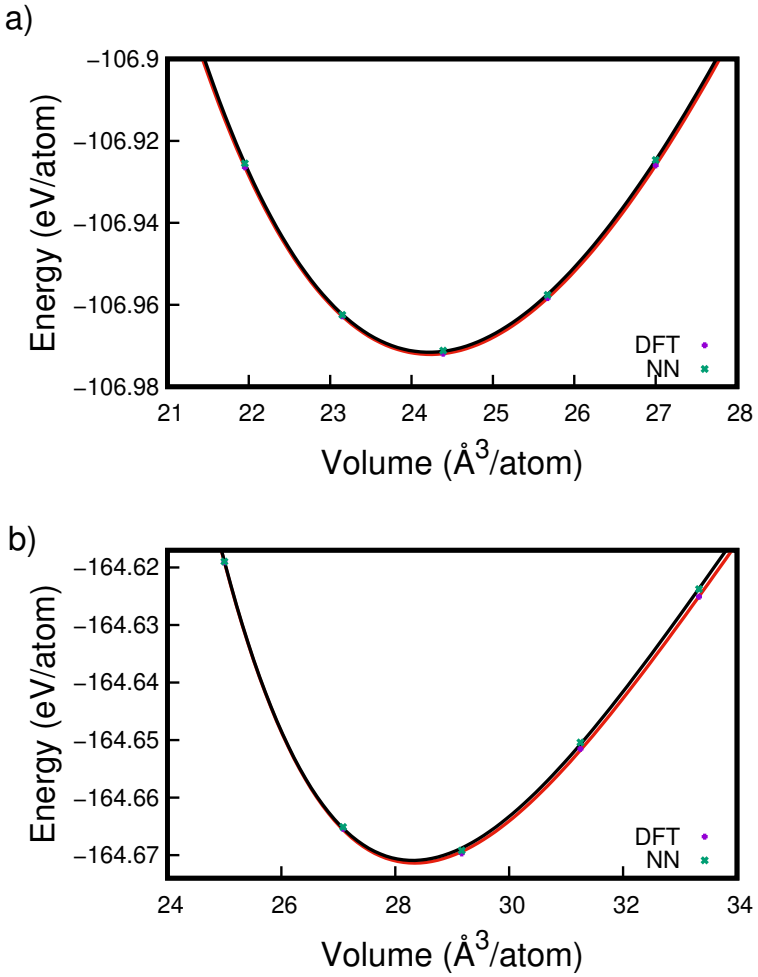

Figure S4: Equation of state from DFT and NN calculations of a) the cubic phase of Ge and b) the trigonal phase of GeTe. The continuous lines are Birch-Murnaghan fit $E(V) = \frac{3B_0}{2}[(V/V_0)^{7/3} - (V/V_0)^{5/3}]\{1 + \frac{3}{4}(B' - 4)[(V/V_0)^{2/3} - 1]$, where V_0 is the equilibrium volume, E_0 the equilibrium energy, B_0 the bulk modulus and B' its derivative respect to pressure. The fitting parameters are given in Table 5 in the article.

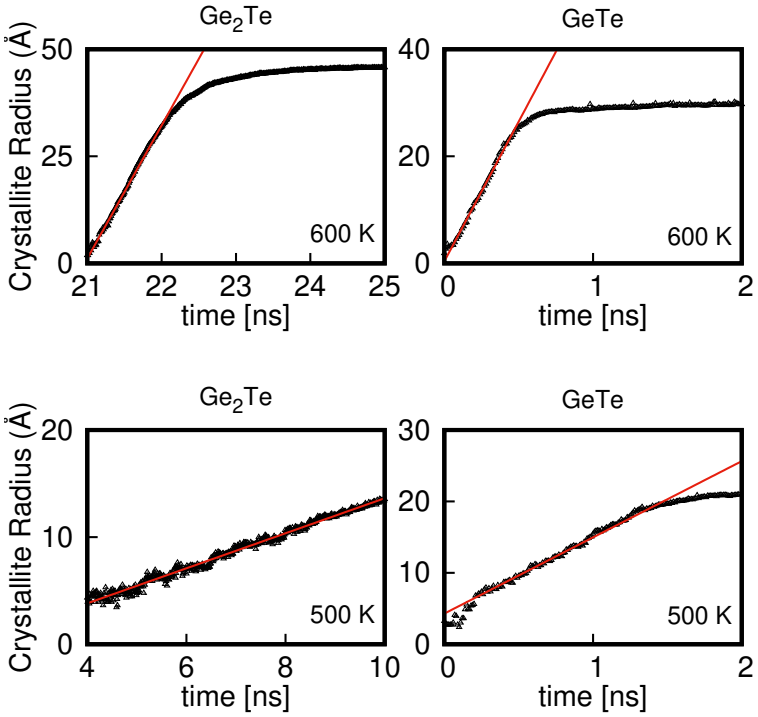

Figure S5: The evolution in time of the radius $R(t)$ of a single GeTe crystalline nucleus in the simulation of (upper panels) stoichiometric GeTe and partially phase separated Ge_2Te at 600 K and (lower panels) stoichiometric GeTe and homogeneous (no phase separation) Ge_2Te at 500 K.

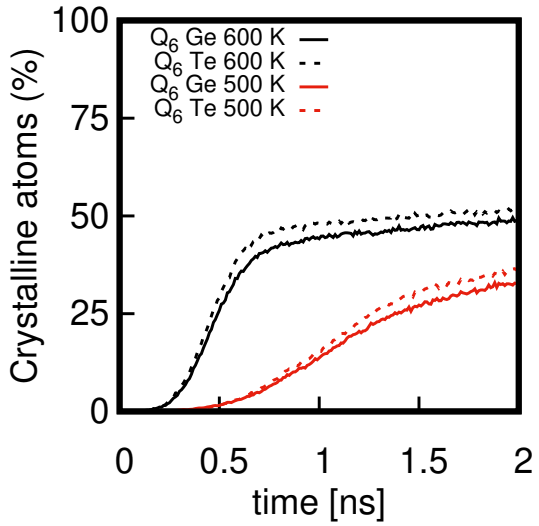

Figure S6: Evolution in time of the Q_6^{dot} crystalline order parameter for the Te (dashed lines) and Ge (continuos lines) fcc sublattices in the crystallization process of GeTe at 600 K and 500 K.

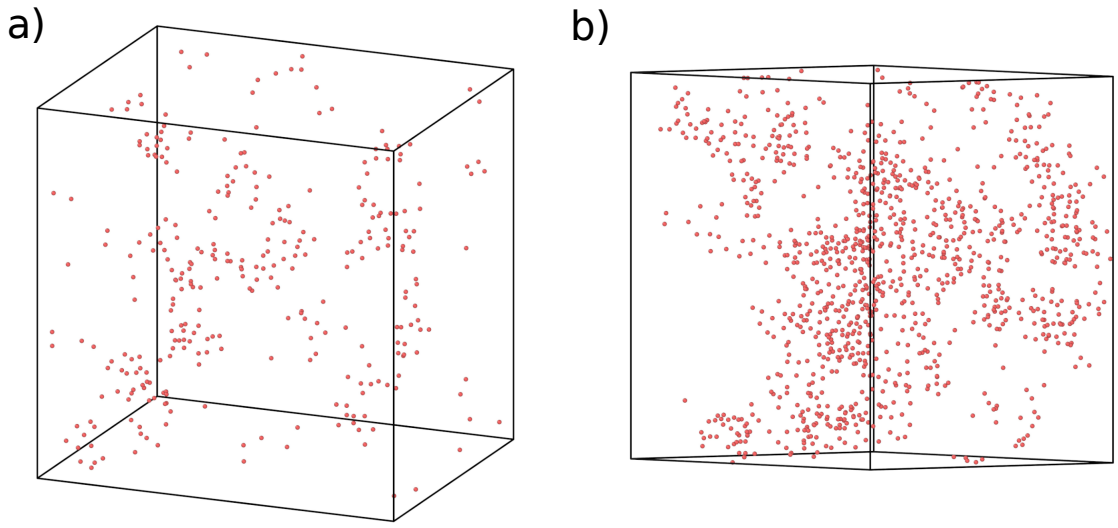

Figure S7: Snapshot of the distribution of Ge atoms with a fraction of Ge-Ge bonds lower than the average value in amorphous GeTe from the simulation of Ge_2Te at a) 500 K and b) 600 K. Crystal nucleation occurs in the regions highlighted in the snapshot.

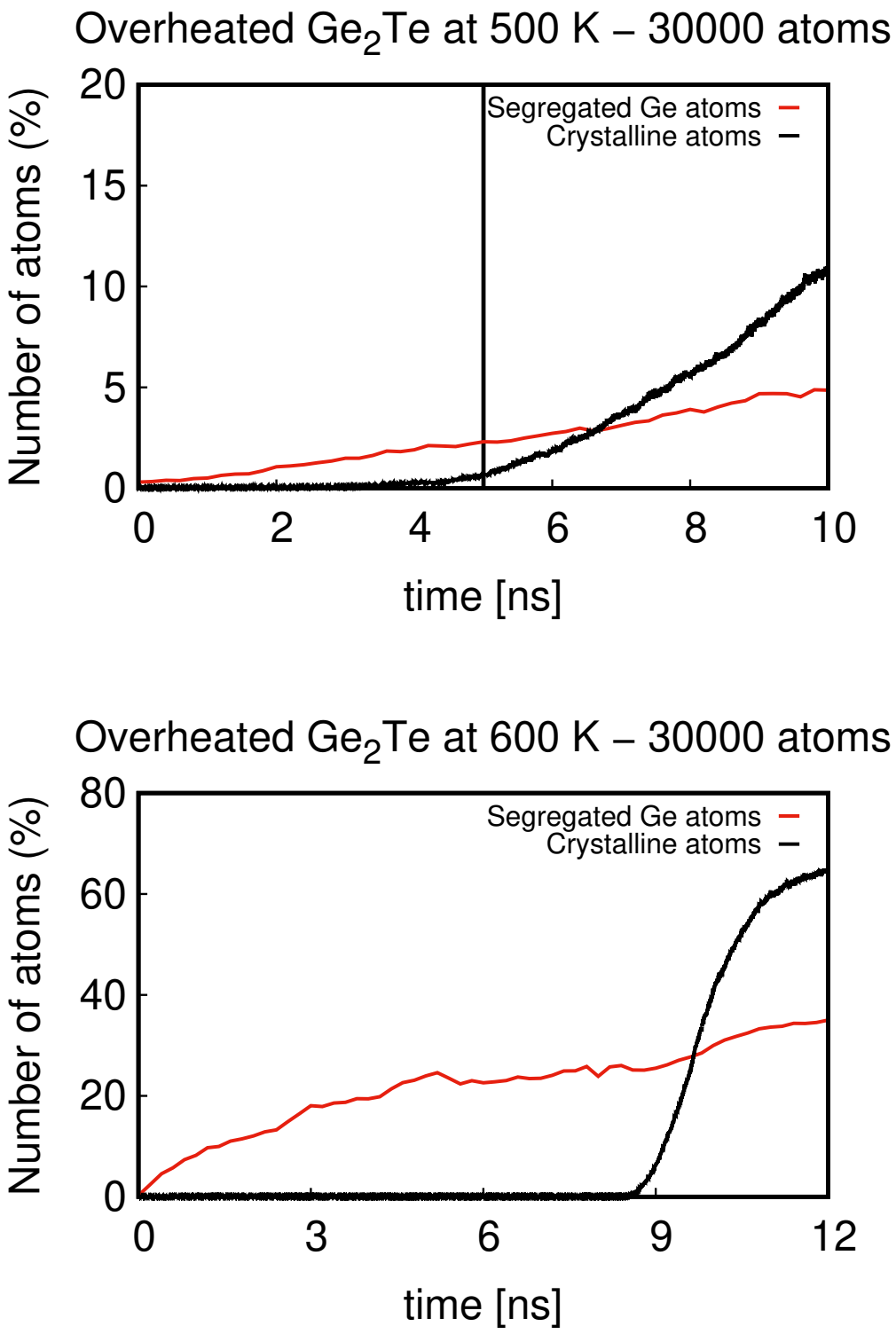

Figure S8: Evolution in time of the number of crystalline atoms and of segregated Ge atoms at 500 K (upper panel) and at 600 K (lower panel) of a 30000-atom model of amorphous Ge_2Te generated by quenching from the melt to 300 K and then annealed at the two target temperatures. The vertical lines indicate the onset of crystallization.

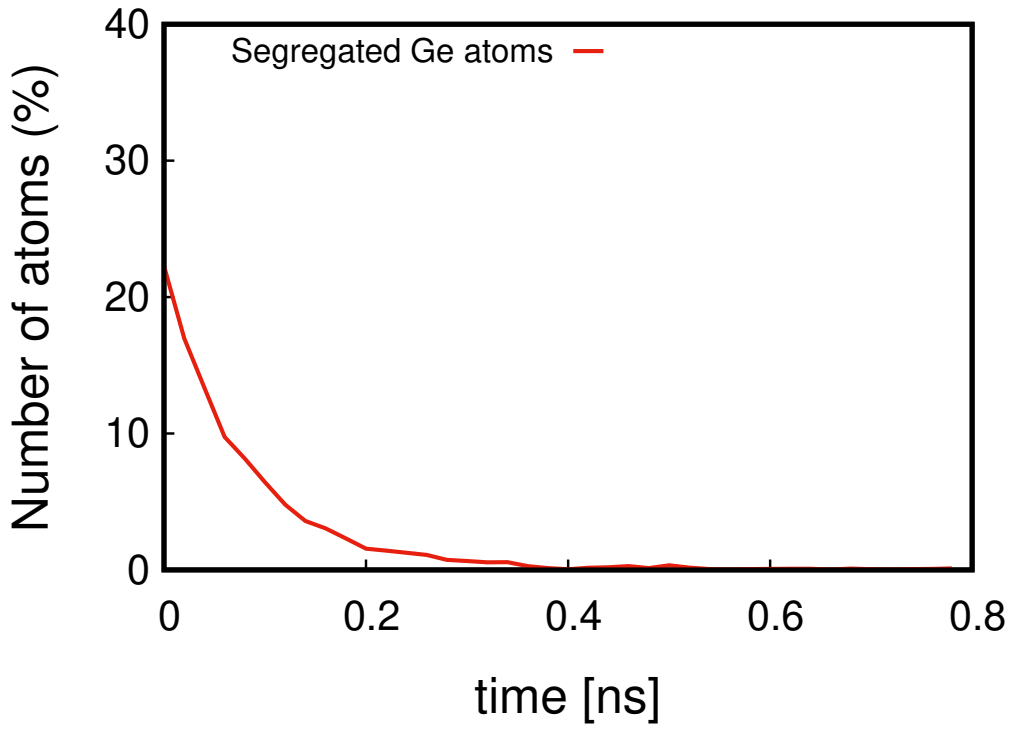

Figure S9: Evolution in time of the number of segregated Ge atoms in the annealing at 1200 K above the liquidus temperature of the phase segregated and crystallized model at 600 K. Before annealing the model was equilibrated at 300 K.