Supporting Information

A Combined ^{77}Se NMR and Molecular Dynamics Contribution to the Structural Understanding of the Chalcogenide Glasses

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Setup of the glass starting configurations

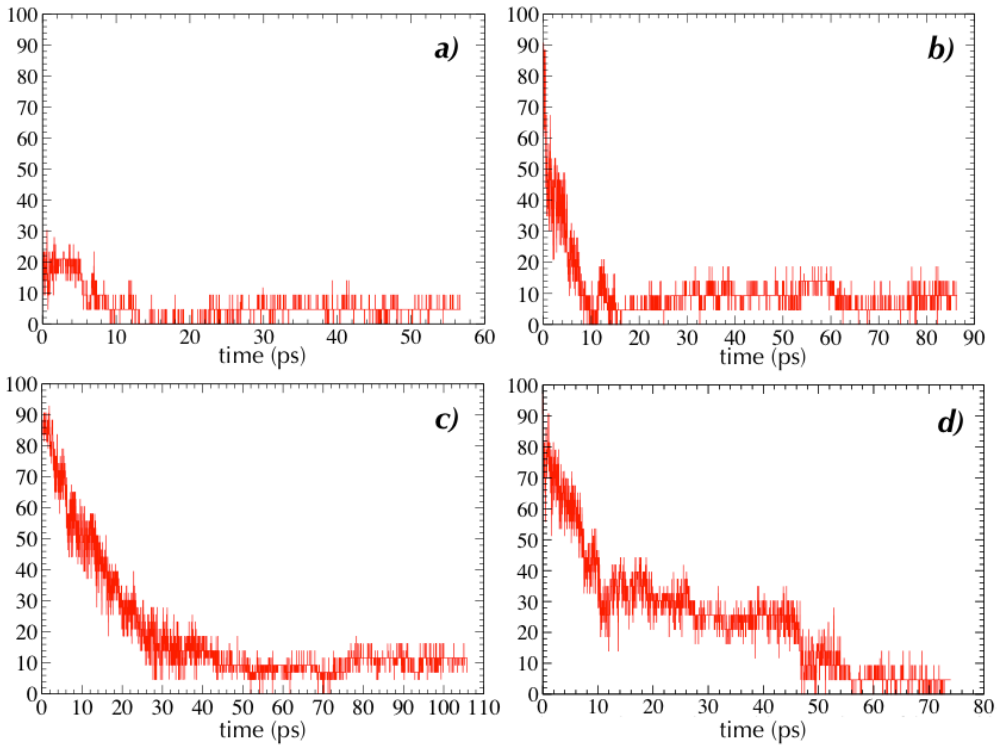

Figure S1. Evolution of the percentage of germanium atoms participating in homopolar bonds during the mixing phase of the different studied configurations. a) – ***H*** structure, b) – ***S*** structure, c) and d) respectively – G_a and G_b structures, corresponding to two independant starting configurations of the ***G*** model.

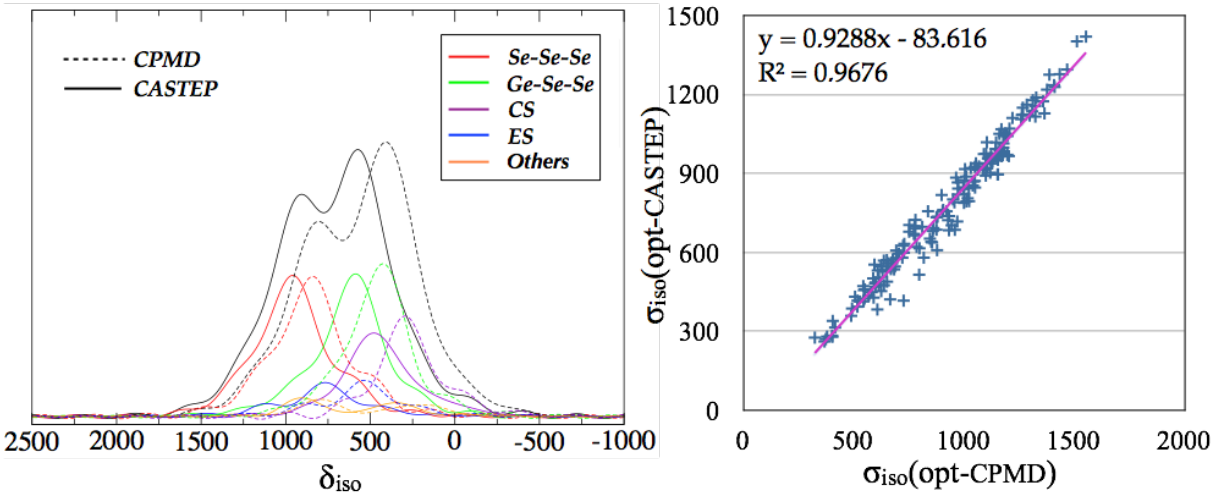

Figure S2. (Left) Simulated ^{77}Se NMR spectra and the various contributions for a configuration taken from the 300K plateau and subsequently optimized with CPMD (dashed lines) and CASTEP (solid lines); (Right) Isotropic chemical shielding values on the CASTEP-optimized structure with respect to the ones of the CPMD-optimized one, together with the least-square fitting.

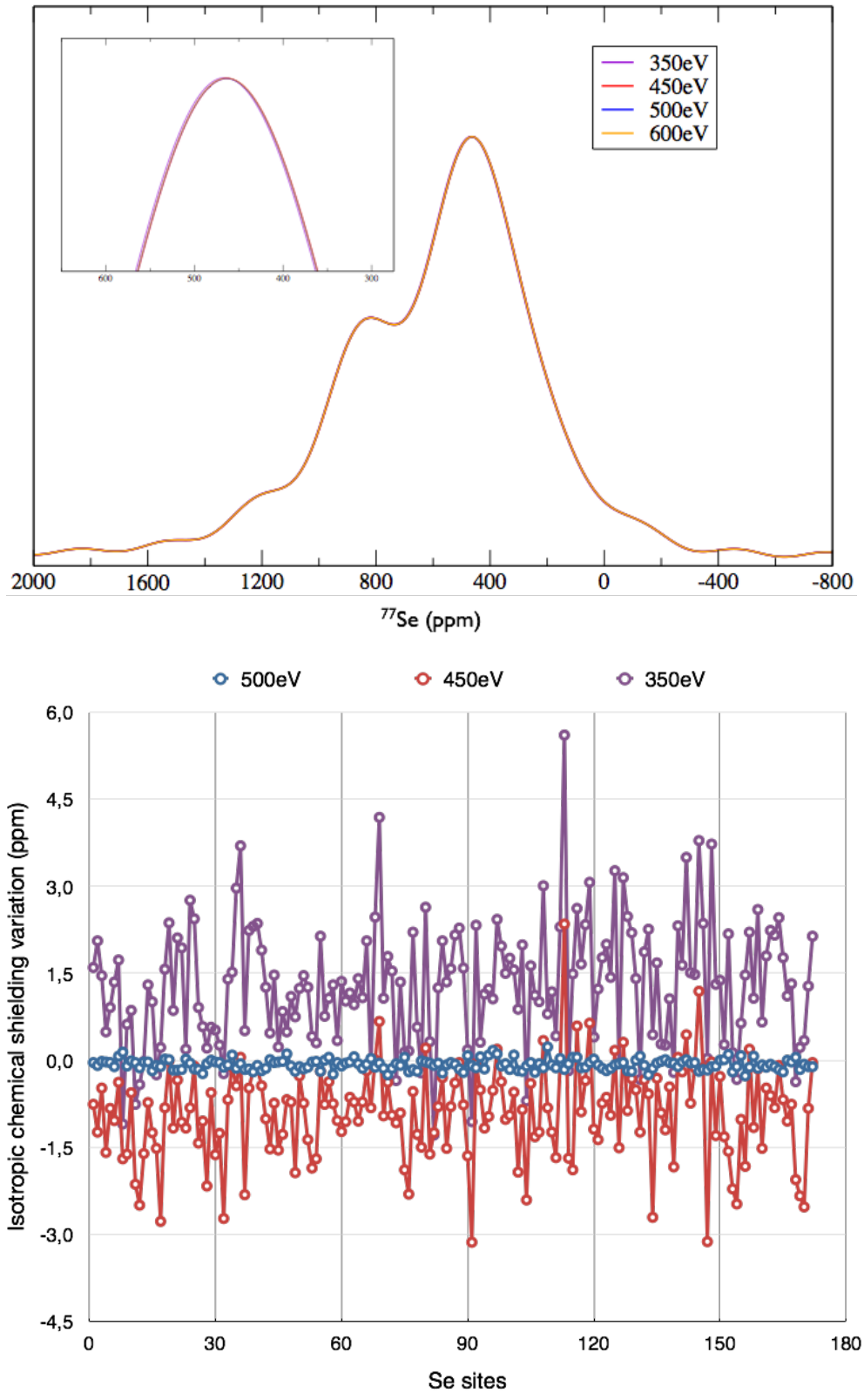

Figure S3. (Top) Simulated ^{77}Se NMR spectra using a 2x2x2 k-points grid and cut-off energies between 350 and 600 eV. The inset contains a zoomed view in the ~300-600 ppm range. (Bottom) Isotropic chemical shielding variations (ppm) with respect to a 600 eV-2x2x2 k-points grid calculation, for each of the 172 Se sites of an optimized glass cell.

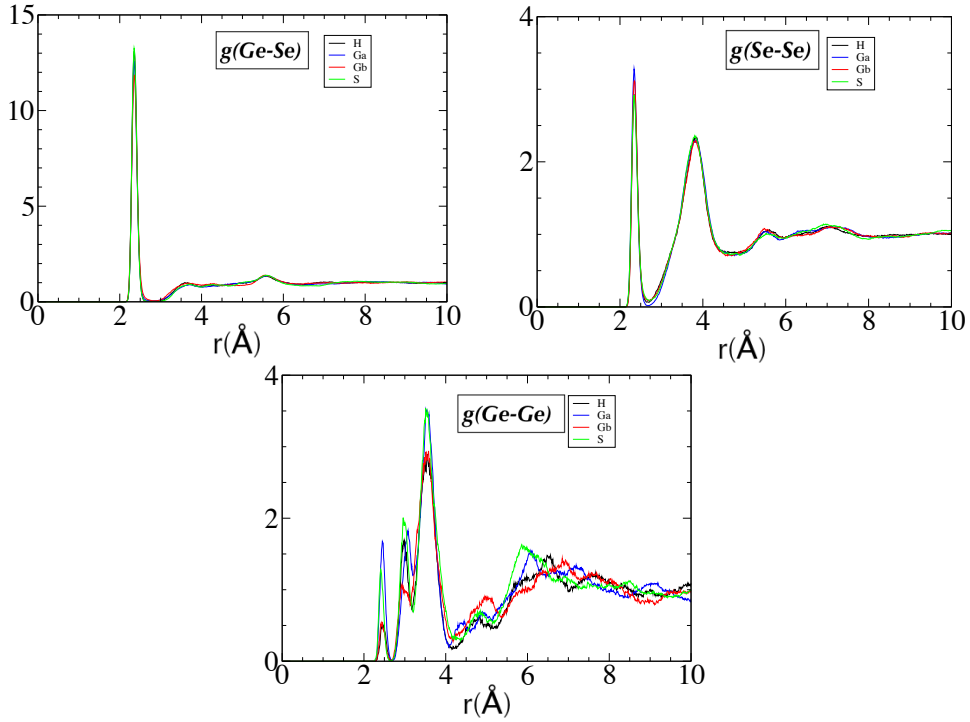

Figure S4. Radial distribution functions calculated for the plateaus at 300 K for ***H*** (black), ***G$_a$*** (blue), ***G$_b$*** (red) and ***S*** (green) models.

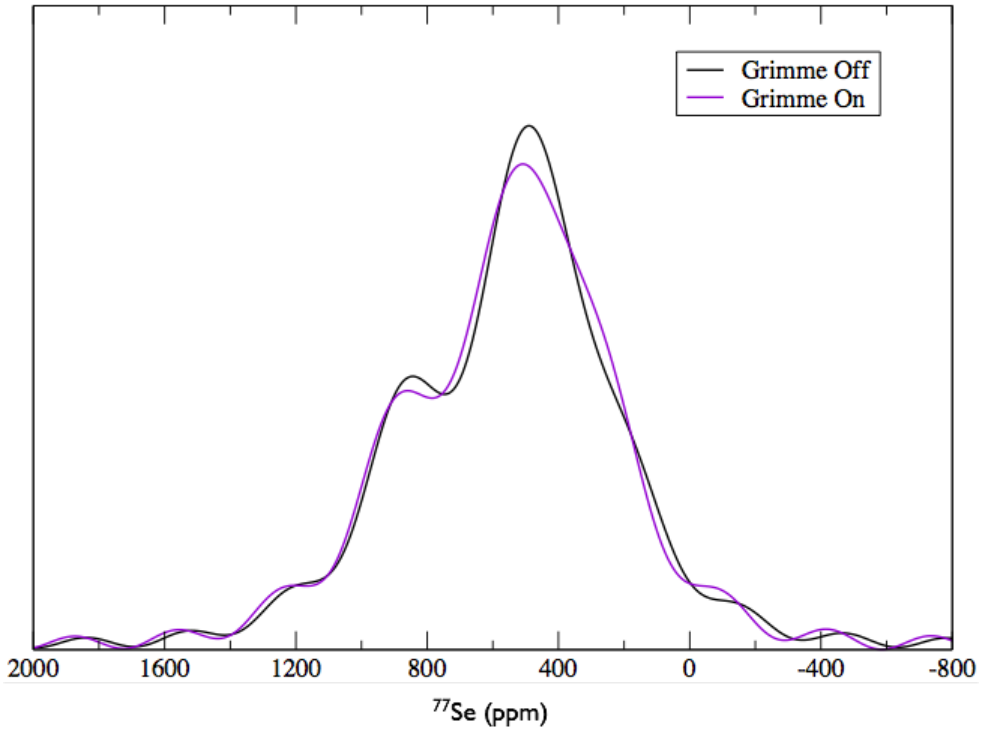

Figure S5. Simulated ^{77}Se NMR spectra on a CPMD-optimized configuration with (black line) and without (violet line) the scheme of Grimme activated during the optimization process.

Setup of the glass starting configurations

The procedure to build up the ***G*** heterogeneous starting configurations relies on a simple two steps process. In a first stage, the germanium atoms are randomly placed in a small volume calculated to correspond approximately to the glass density. In a second stage, this germanium grain is positioned in the center of an empty cell whose dimensions correspond to the ones of the final glass cell. Selenium atoms are subsequently inserted randomly within the empty space of the cell. During the two stages, a minimum distance criterion is used in order to avoid excessive heating of the glass during the equilibration phase in the liquid state. Two different cells ($\boldsymbol{G_a}$ and $\boldsymbol{G_b}$) have been built up using this random process corresponding therefore to different sets of initial positions for both the germanium and selenium phases.