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Fig. S1: Electron microscope images of A$\beta_{1\text{-}40}$ fibrils prepared as described in "Methods" section. a) the batch used to prepare frozen solution Aβ-VF fibrils; b) batch used to prepare frozen solution Aβ-VG fibrils.

Fig. S2: DNP build-up curves measured using proton signals and ^{13}C-CP signals, monoexponential fitting curves are shown in red. a) Aβ-VF fibrils in frozen solution, b) Aβ-VF peptide in frozen solution, c)Aβ-VG fibrils in frozen solution, d) transverse relaxation curves for samples in frozen solution.

Fig. S3: Schematic representation of relative orientation of CSA tensors in the molecular frame and with respect to each other.

Fig. S4: Series of simulated 2D patterns for a wide range of various (ϕ,ψ) angles. Spectral broadening is set to match the one found in room temperature measurements, exchange time 2.5 s

Fig. S5: $\chi^2_{red}(\phi,\psi)$ plots for the best fits to individual 2D exchange spectra of Aβ-VF fibrils with various $\tau_{exchange}$. (data are shown in Fig. 6), a) $\tau_{exchange}$.=0.1 s, b) $\tau_{exchange}$.=0.5 s, c) $\tau_{exchange}$.=1.0 s, d) $\tau_{exchange}$.=2.5 s, e) $\tau_{exchange}$.=10 s.

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Fig. S6: RMSD plot for simulated data. The RMSD is calculated as $RMSD(\phi,\psi) = \sum_{i}^{N}(\lambda S_i(\phi,\psi) - \lambda S_i(\phi_0,\psi_0))^2$, where a spectrum S_i simulated for specific (ϕ_0,ψ_0) angles is compared to all other simulations, the optimal scaling factor l is calculated as $\lambda = \frac{\sum_i^N S_i(\phi_0,\psi_0)S_i}{\sum_i^N S_i(\phi_0,\psi_0)S_i}$ a) (ϕ_0,ψ_0) =(-50º,-60º) ; b) (ϕ_0,ψ_0) =(-120º,120º); c) (ϕ_0,ψ_0) =(-150º,150º); d) (ϕ_0,ψ_0) =(-75º,-150º)

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