Supporting Information

Perturbation of Long-Range Water Dynamics as the Mechanism for the Antifreeze Activity of Antifreeze Glycoprotein.

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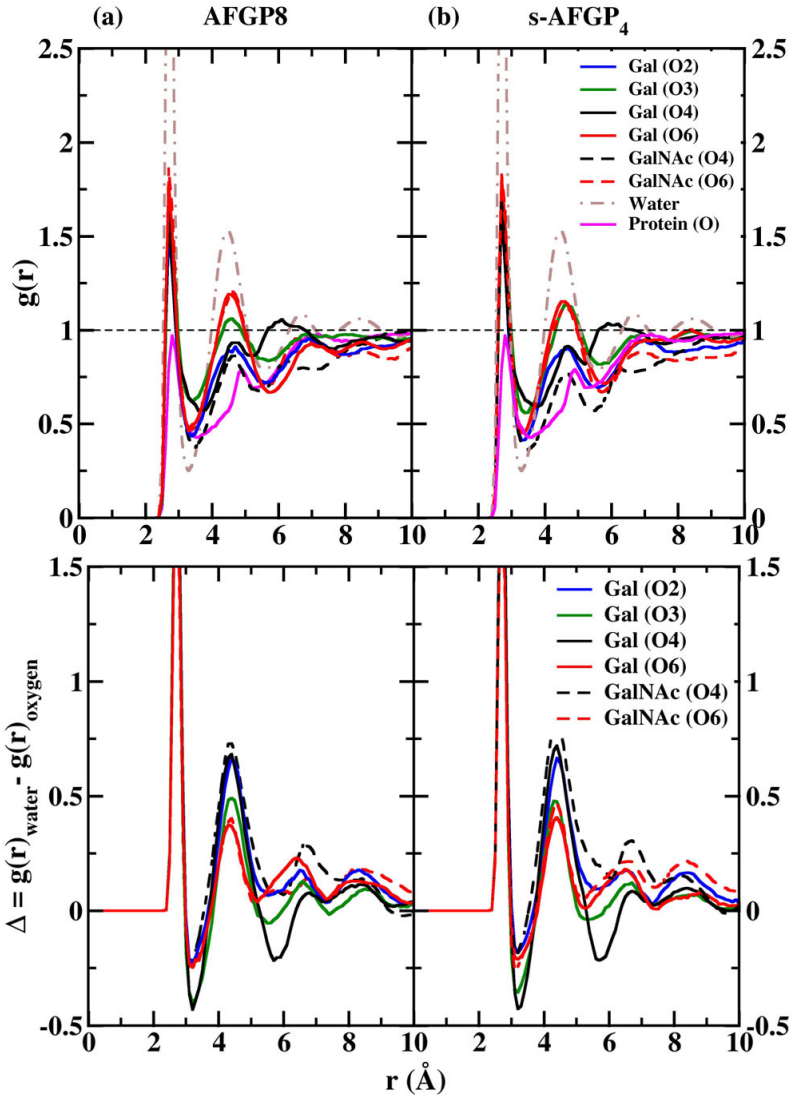

Figure S1: Upper Panel: Selected oxygen (carbohydrate) – oxygen (water) radial distribution functions, g(r), from 250K simulations of (a) AFGP8 and (b) s-AFGP$_4$ using the TIP5P water model. The g(r) for pure water (TIP5P water model) under the same simulation conditions is also presented for comparison. **Lower Panel:** Difference (Δ) between the radial distribution functions. Δ is calculated as the difference between g(r) of pure water and g(r) of select oxygen (carbohydrate) -oxygen (water) radial distribution from 250K simulations of (a) AFGP8 and (b) s-AFGP$_4$.

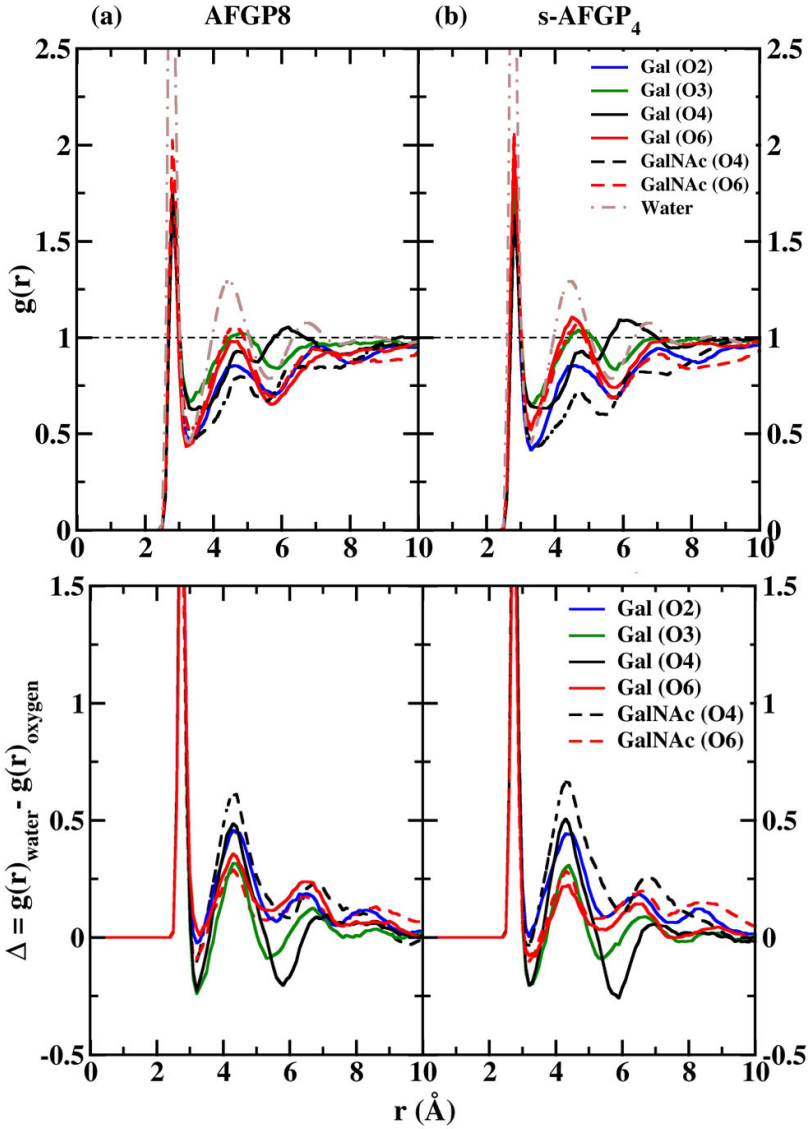

Figure S2: Upper Panel: Selected oxygen (carbohydrate) – oxygen (water) radial distribution functions, g(r), from 250K simulations of (a) AFGP8 and (b) s-AFGP$_4$ using the TIP4P-2005 water model. The g(r) for pure water (TIP4P-2005 water model) under the same simulation conditions is also presented for comparison. **Lower Panel:** Difference (Δ) between the radial distribution functions. Δ is calculated as the difference between g(r) of pure water and g(r) of select oxygen (carbohydrate) -oxygen (water) radial distribution from 250K simulations of (a) AFGP8 and (b) s-AFGP$_4$.

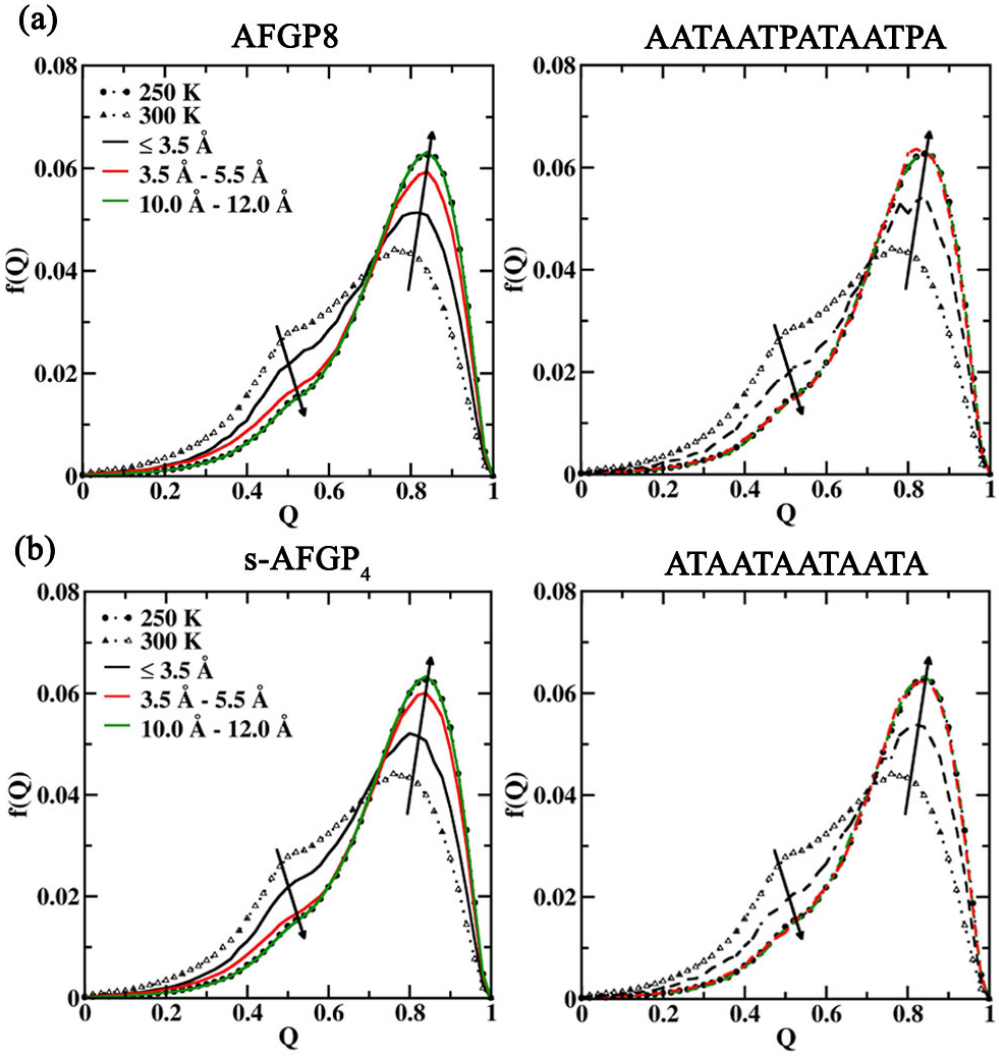

Figure S3: Q distributions for water molecules around Gal (left panels) from 250 K simulations of (a) AFGP8 and (b) s-AFGP$_4$. The Q distribution for water molecules around protein (right panels) from 250 K protein-only simulations of (a) AATAATPATAATPA and (b) ATAATAATAATA are also presented. The distributions were evaluated for water molecules less than 3.5 Å, between 3.5 Å-5.5 Å, and between 10.0 Å-12.0 Å from the selection (Gal or protein). The Q distribution for pure water at 250 K (dotted line with filled circle) and 300 K (dotted line with open triangles) are also presented. The distributions are evaluated from simulations using the TIP4P-2005 water model.

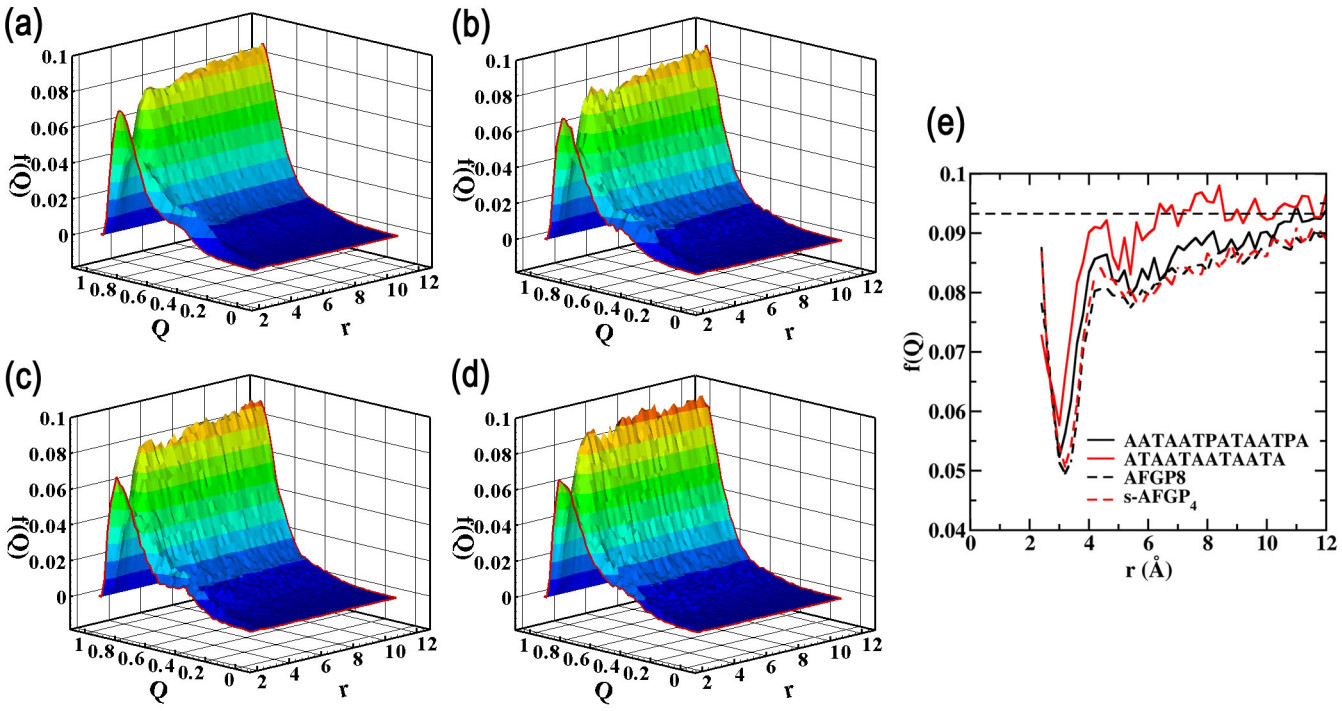

Figure S4: Q_K distribution for water molecules around Gal evaluated in increments of 0.2 Å shells from the glycopeptides from 250 K simulations of (a) AFGP8 and (b) s-AFGP$_4$. The Q_K distribution for water molecules around protein evaluated in increments of 0.2 Å shells from 250 K protein-only simulation of (c) AATAATPATAATPA and (d) ATAATAATAATA. (e) The trace of the highest Q_k peak from all these profiles. Dotted line indicates the value of the highest Qk peak from pure water simulations at 250 K. The distributions are evaluated from simulations using the TIP5P water model.

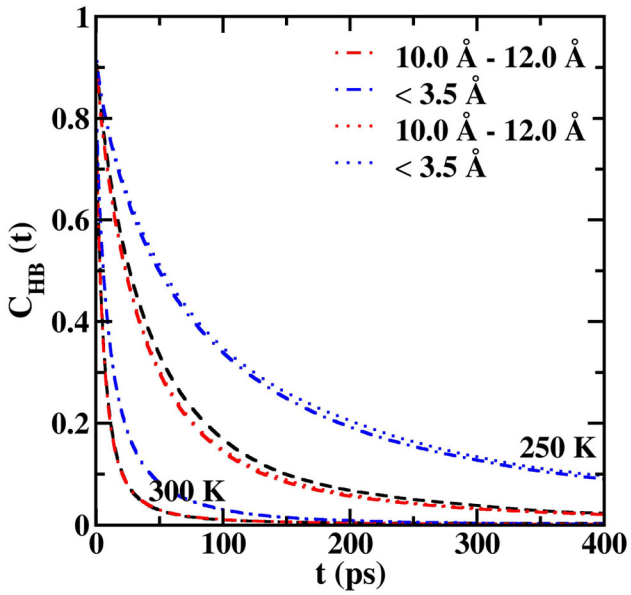

Figure S5: Water-water H-bond autocorrelation functions from 250 K and 300 K simulations of AFGP8 (dot dashed lines) and s-$AFGP_4$ (dotted lines). The autocorrelation functions were evaluated for water molecules within two spatial regions, less than 3.5 Å (blue lines) and 10.0 Å -12.0 Å (red lines) from the glycopeptides. Autocorrelation functions evaluated from 250 K and 300 K pure water simulations (dashed black line) are also presented for comparison. The distributions are evaluated from simulations using the TIP4P-2005 water model.

AFGP8		s-$AFGP_4$		Water	
250 K	300 K	250 K	300 K	250 K	300 K
TIP5P					
0.0066	0.3011	0.0066	0.2998	0.0047	0.2778
TIP4P-2005					
0.0367	0.2253	0.0362	0.2248	0.0332	0.2137

Table S1: Diffusion Coefficient (D) estimated from the mean square displacement (MSD) of water oxygen atoms. MSD were evaluated from the last 2 ns of the molecular dynamics simulations used to calculate the H-bond auto-correlation functions.