Supporting Table 2. Calculated model-free parameters for the tetra-, hexa- and octa-saccharides (**1**, **2** and **3** respectively) of the NA-domain of heparan sulfate, using a constant internal correlation time of 30 ps, fitted to isotropic and anisotropic models of rotational diffusion. These values were calculated from experimental data collected at three magnetic-field strengths (14.1, 18.1 and 21.1 T). The S^2_{calc} and α_{calc} values were calculated by averaging data extracted from 50 ns molecular dynamics simulations. In contrast to Table 2 (where a value of 1.01Å was used for the N-H distance) a value of 1.02Å was used for the N-H distance in model free analysis.

Isotropic			S^2	S^2_{calc}
1				
τ_M [ns]	0.55	α	0.35	0.46
τ_e [ns]	0.03	β	0.38	0.59
		ω	0.52	0.62
2				
τ_M [ns]	0.68	α	0.37	0.49
τ_e [ns]	0.03	β	0.39	
		γ	0.64	0.70
		ω	0.59	0.61
3				
τ_M [ns]	0.74	α	0.35	
τ_e [ns]	0.03	β	0.38	
		γ	0.93	
		ψ	0.88	
		ω	0.74	

(see over for anisotropic data).

Anisotropic			S^2	α[°]	S^2_{calc}	α_{calc}[°]
1						
$\tau_{\parallel}$ [ns]	0.35	α	0.32	7.3	0.46	64.2
$\tau_{\perp}$ [ns]	0.63	β	0.39	42.3	0.59	62.1
$\tau_{\perp}/\tau_{\parallel}$	1.77	ω	0.55	57.5	0.62	63.1
τ_e [ns]	0.03					
2						
$\tau_{\parallel}$ [ns]	0.56	α	0.33	5.5	0.49	62.8
$\tau_{\perp}$ [ns]	0.75	β	0.39	54.0		
$\tau_{\perp}/\tau_{\parallel}$	1.34	γ	0.66	57.3	0.70	64.4
τ_e [ns]	0.03	ω	0.65	85.7	0.61	63.3
3						
$\tau_{\parallel}$ [ns]	0.38	α	0.42	72.3		
$\tau_{\perp}$ [ns]	0.98	β	0.45	65.2		
$\tau_{\perp}/\tau_{\parallel}$	2.57	γ	0.68	2.5		
τ_e [ns]	0.03	ψ	0.70	23.0		
		ω	0.58	32.0		