Supplementary information: Figure S1-S11 and Table S1

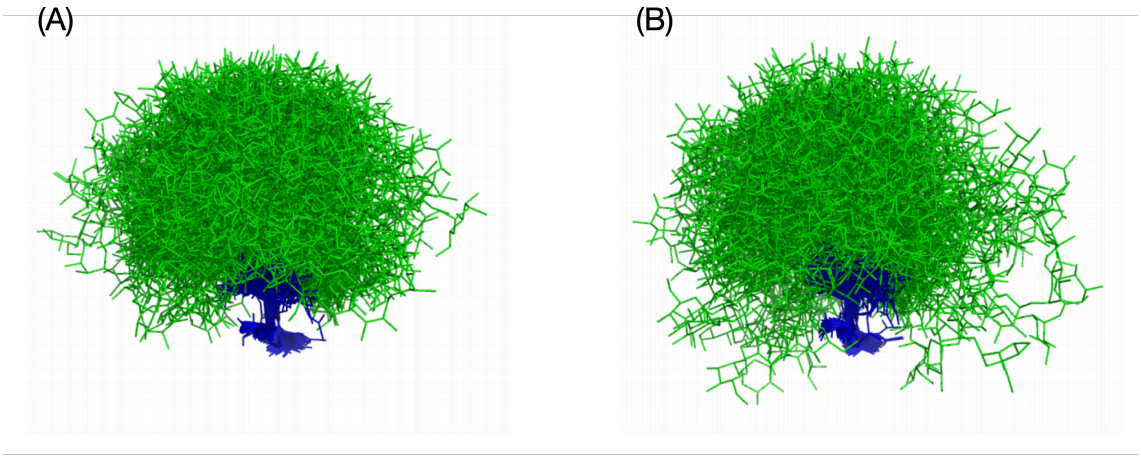

Fig. S1: Conformational ensembles of the high-mannose-type oligosaccharides (A) M9 and (B) M8B. The ensemble data were obtained from the replica-exchange MD calculation in the previous study13. The GlcNAc and mannose residues are represented by blue and green, respectively.

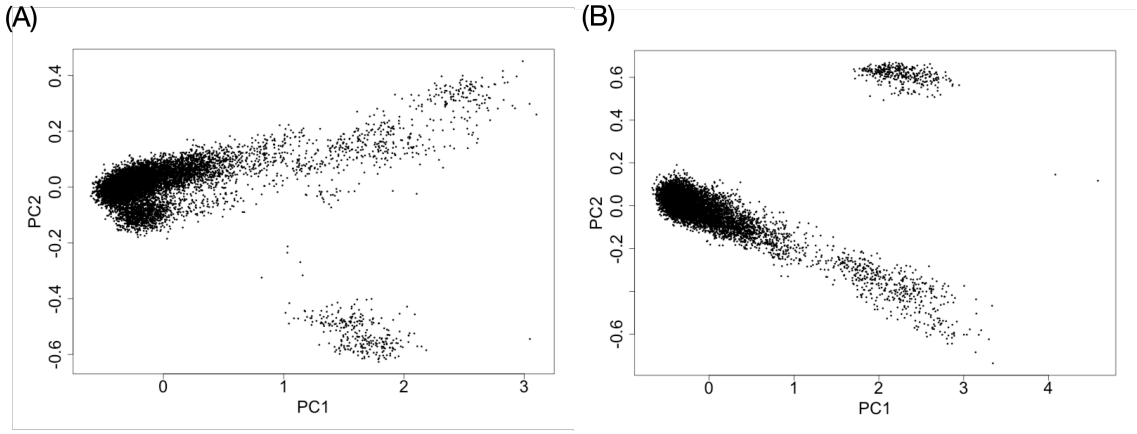

Fig. S2: The scatter plots of the first two kernel principal components for RKHS projected from the conformational ensemble data of (A) M9 and (B) M8B.

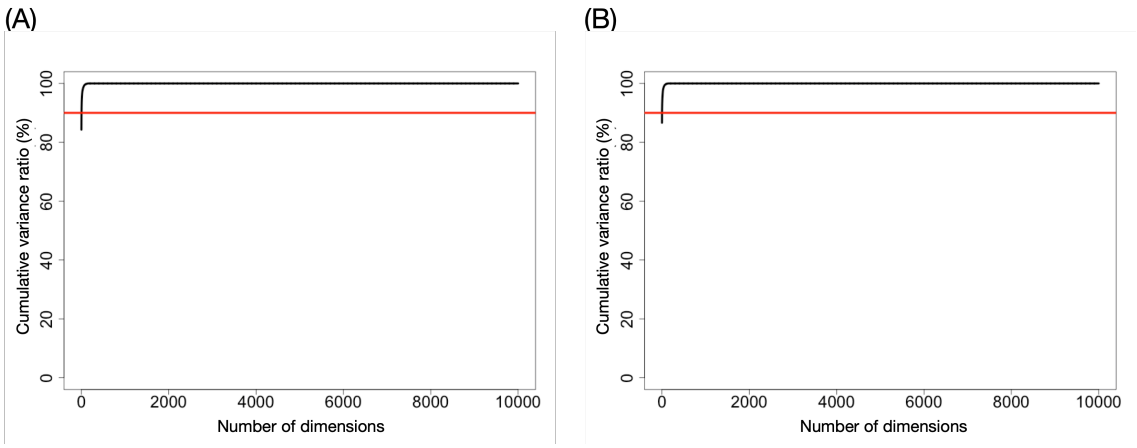

Fig. S3: Relationship between the number of dimensions of RKHS and its cumulative proportion. RKHS was projected from the conformational ensemble data of (A) M9 and (B) M8B. The horizontal red line shows 90 % of the cumulative proportion.

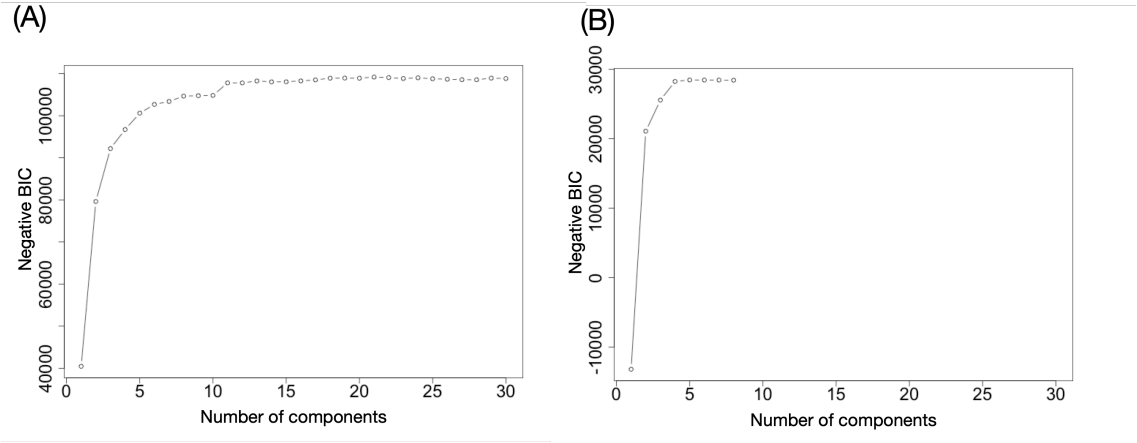

Fig. S4: The BIC values corresponding to different numbers in RKHS of (A) M9 and (B) M8B. The vertical axes show negative BIC values.

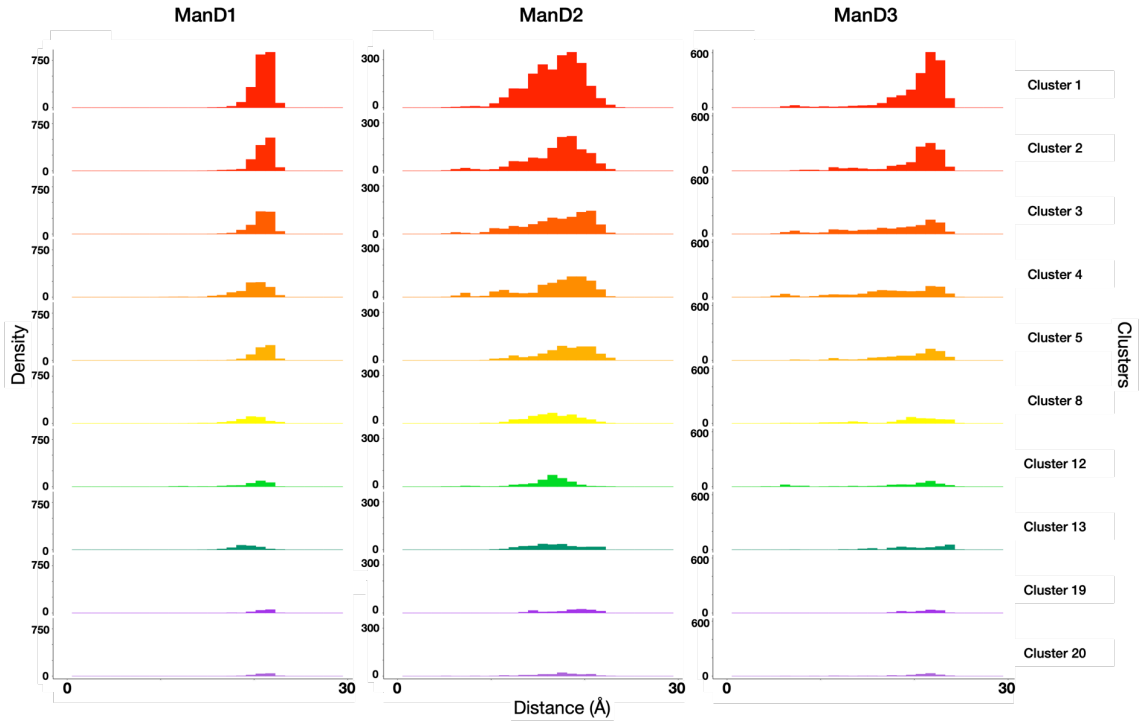

Fig S5: Histograms of the distances between the anomeric carbons of the reducing-terminal GlcNAc1 residue and each outer mannose residues of the 10 kinds of M9 clusters classified as extended forms.

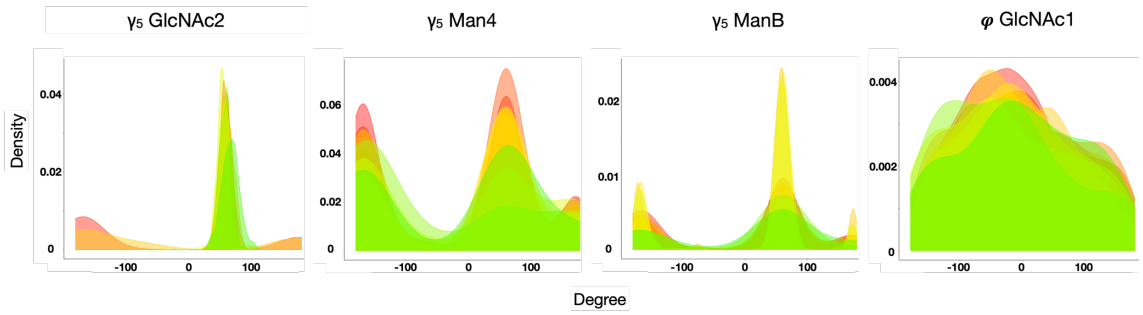

Fig. S6: The distribution profiles of the orientation of hydroxyl groups γ_5 in GlcNAc2, Man4 and ManB and the ring puckering states $\boldsymbol{\varphi}$ in GlcNAc1 among the 10 kinds of M9 clusters classified as extended forms.

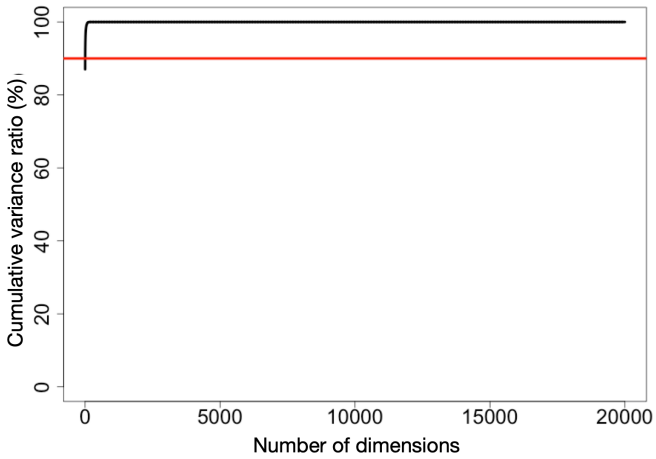

Fig S7: Relationship between the number of dimensions of common RKHS in M9* and M8B and its cumulative proportion. The common RKHS was projected from the conformational ensemble data derived from the MD simulations of M9* and M8B. The horizontal red line shows 90 % of the cumulative proportion.

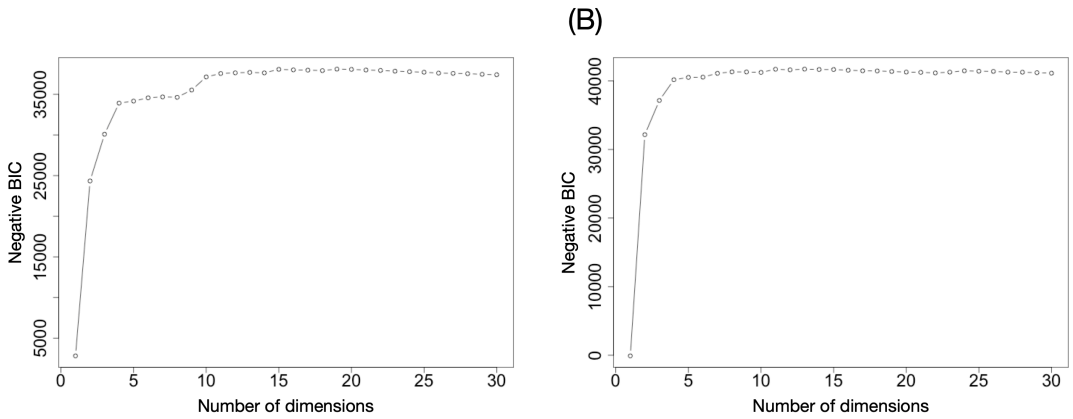

Fig. S8: The BIC values corresponding to different numbers in common RKHS of (A) M9* and (B) M8B. The vertical axes show negative BIC values.

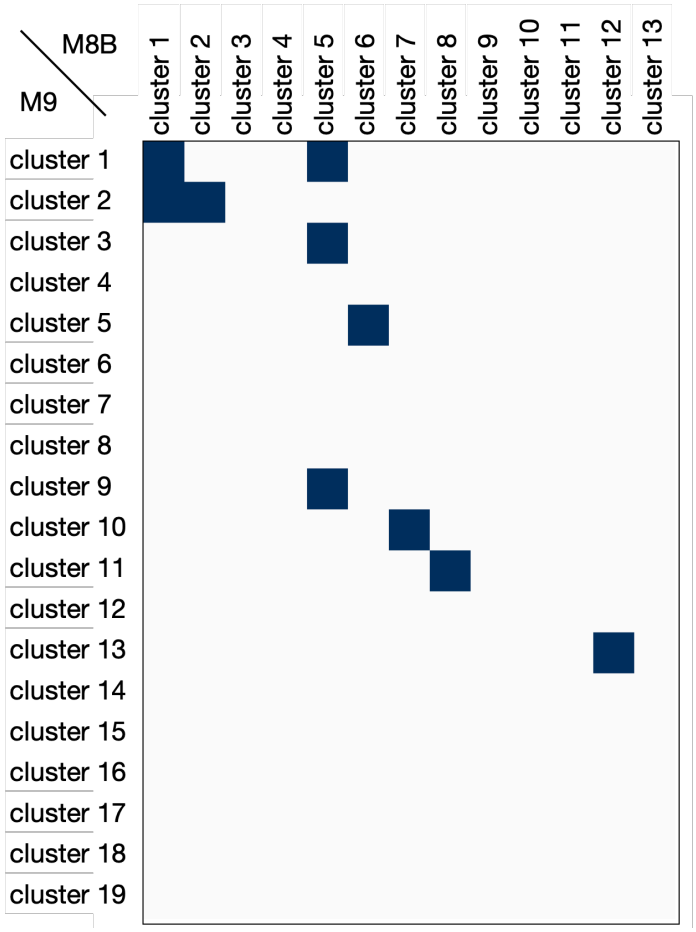

Fig. S9: Matrix-like plot for identification of M9*- and M8B-characteristic clusters. The cells are filled when MMD values between the M9* and M8B clusters is below the threshold of 0.100.

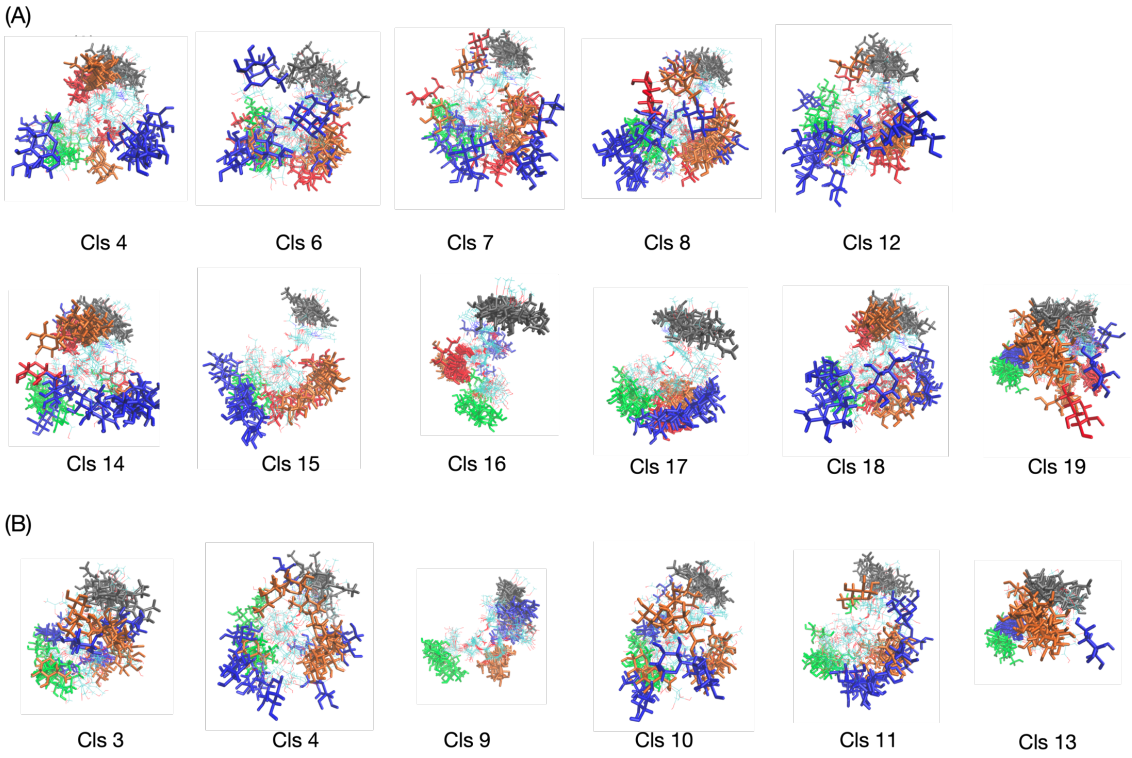

Fig. S10: Molecular graphical representation of the M8B- and M9*-characteristic conformational clusters. The superimposition of 10 structures were extracted from each of (A) 11 of M9*-characteristic clusters and each of (B) 6 of M8B-characteristic clusters. These clusters occupy more than 1 % of all ensemble populations. The ManD1, Man D2, MaD3, ManA, and GlcNAc1 residues are colored in green, red, blue, yellow, and black, respectively.

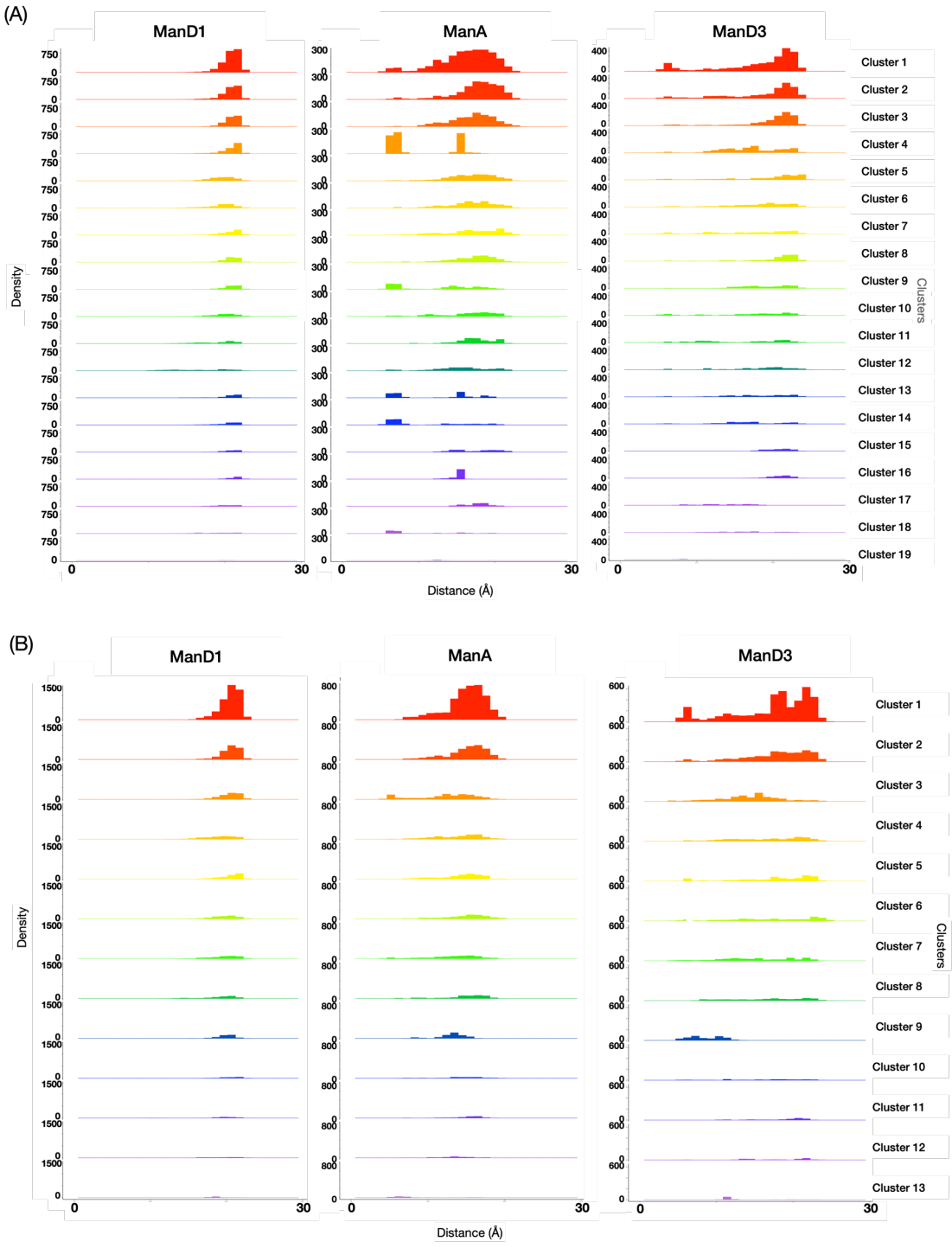

Fig. S11: Histograms of the distances between the anomeric carbons of the reducing-terminal GlcNAc1 residue and each outer mannose residue of the19 and 13 kinds of (A) M9* and (B) M8B clusters, respectively.

Table S1: The actual values of scaling parameters and hyperparameters used in the kernel analysis for characterizing the M9 and M8B ensembles as well as comparing the M9* and M8B ensembles.

	M9	M8B	M9* and M8B
σ (hyperparameter)	98.98238	89.8572	90.46392
φ GlcNAc1	0.74307709	0.74431556	0.74119813
φ GlcNAc2	0.75124586	0.75503287	0.74034606
φ Man3	0.70377374	0.67454955	0.69422538
φ Man4	0.69739102	0.67906888	0.69169428
φ Man4'	0.65062515	0.69060820	0.67311928
φ ManA	0.69726916	0.64930564	0.67064742
φ ManB	0.67707432	0.67989609	0.68128401
φ ManC	0.69450344	0.67631405	0.68073445
φ ManD1	0.67884033	0.68039850	0.67895258
φ ManD2	0.65535527	NA	NA
φ ManD3	0.65923318	0.64585911	0.65017101
θ GlcNAc1	0.08027921	0.07884240	0.08019819
θ GlcNAc2	0.07935273	0.08047762	0.07993478
θ Man3	0.07654154	0.07667769	0.07670820
θ Man4	0.07236693	0.07172492	0.07209811
θ Man4'	0.07463972	0.07662912	0.07538426
θ ManA	0.07323564	0.07246493	0.07235141
θ ManB	0.07345467	0.07403877	0.07426858
θ ManC	0.07193312	0.07212442	0.07168355
θ ManD1	0.07641618	0.07039497	0.07271084
θ ManD2	0.07037427	NA	NA
θ ManD3	0.07213393	0.07088933	0.07118775
ϕ GlcNAc2-GlcNAc1	0.16480597	0.16924334	0.16758437
ϕ Man3-GlcNAc2	0.20971947	0.23324450	0.22437981
ϕ Man4-Man3	0.16554207	0.16828220	0.17028939
ϕ Man4'-Man3	0.20353548	0.22209233	0.21547123
ϕ ManA-Man4'	0.26899157	0.17569795	0.20834682
ϕ ManB-Man4'	0.19154701	0.19025109	0.19060621
ϕ ManC-Man4	0.16673160	0.16443163	0.16645672
ϕ ManD1-ManC	0.15951205	0.15932030	0.16001677
ϕ ManD2-ManA	0.13896695	NA	NA
ϕ ManD3-ManB	0.16085879	0.16079284	0.16011206

ψ GlcNAc2-GlcNAc1	0.28218793	0.28207401	0.28206336
ψ Man3-GlcNAc2	0.25514565	0.32416342	0.29355212
ψ Man4-Man3	0.47470700	0.45195420	0.46737082
ψ Man4'-Man3	0.33187813	0.55371395	0.42621633
ψ ManA-Man4'	0.44047788	0.43841041	0.44157795
ψ ManB-Man4'	0.68762230	0.81896991	0.78545558
ψ ManC-Man4	0.45265030	0.45751623	0.44792605
ψ ManD1-ManC	0.42350124	0.41656446	0.42159736
ψ ManD2-ManA	0.34976709	NA	NA
ψ ManD3-ManB	0.44151897	0.41631929	0.42457184
ω Man4'-Man3	0.61671929	0.86294334	0.75679833
ω ManB-Man4'	1.42461301	1.50340001	1.49142767
γN GlcNAc1	0.30603109	0.31168878	0.30239925
γN GlcNAc2	0.27523828	0.28211591	0.27305772
γO GlcNAc1	0.16420597	0.16827665	0.16795172
γO GlcNAc2	0.16286132	0.16215802	0.16383678
γ2 Man3	1.08081347	0.90389884	1.02281131
γ2 Man4'	0.99932948	1.02990890	1.02412187
γ2 ManD1	0.84949774	0.79429176	0.80355172
γ2 ManD2	0.71496921	NA	NA
γ2 ManD3	0.78876946	0.84914814	0.84884201
γ3 GlcNAc1	0.46715143	0.43449217	0.45203970
γ3 GlcNAc2	0.52192276	0.70333910	0.60930939
γ3 Man4	1.41922189	1.42414322	1.43483709
γ3 ManA	1.43773999	1.42770975	1.43881613
γ3 ManB	1.39125531	1.41485031	1.40223166
γ3 ManC	1.42065388	1.42663416	1.45205330
γ3 ManD1	1.39713621	1.39783784	1.41247446
γ3 ManD2	1.39593145	NA	NA
γ3 ManD3	1.41785375	1.43020628	1.42478943
γ4 Man3	0.53074126	0.55750598	0.55378031
γ4 Man4	0.67685435	0.71386897	0.69513155
γ4 Man4'	0.69600392	0.60728014	0.63975967
γ4 ManA	0.70405166	0.70877133	0.71744013
γ4 ManB	0.73119916	0.75018222	0.73365300
γ4 ManC	0.75638930	0.79948010	0.74901063
γ4 ManD1	0.79909604	0.74207531	0.77777616
γ4 ManD2	0.74483610	NA	NA

γ4 ManD3	0.80451468	0.76626470	0.78534809
γ5 GlcNAc1	1.11987885	0.75634917	0.86230830
γ5 GlcNAc2	0.35257172	0.53271522	0.40577036
γ5 Man4	1.15798535	1.19315618	1.12906324
γ5 ManA	1.19687755	1.21255960	1.10129533
γ5 ManB	1.06169298	0.98252664	1.07448410
γ5 ManC	1.24211052	1.15105057	1.09462906
γ5 ManD1	1.13337685	1.04346149	1.09076855
γ5 ManD2	1.25750806	NA	NA
γ5 ManD3	1.22071424	1.12077533	1.26753647
γ6 GlcNAc1	1.38225537	1.33646778	1.35036733
γ6 GlcNAc2	1.27295117	1.20096272	1.23995324
γ6 Man4	1.42874247	1.42661846	1.40605222
γ6 ManA	1.46779708	1.47978532	1.45249059
γ6 ManB	1.37472914	1.37253886	1.38459529
γ6 ManC	1.47605055	1.44288712	1.45580554
γ6 ManD1	1.43623095	1.43147908	1.44865572
γ6 ManD2	1.45961254	NA	NA
γ6 ManD3	1.45532192	1.45625967	1.46250309