Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information

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I. Section S1. Additional Details of Atomistic Simulations

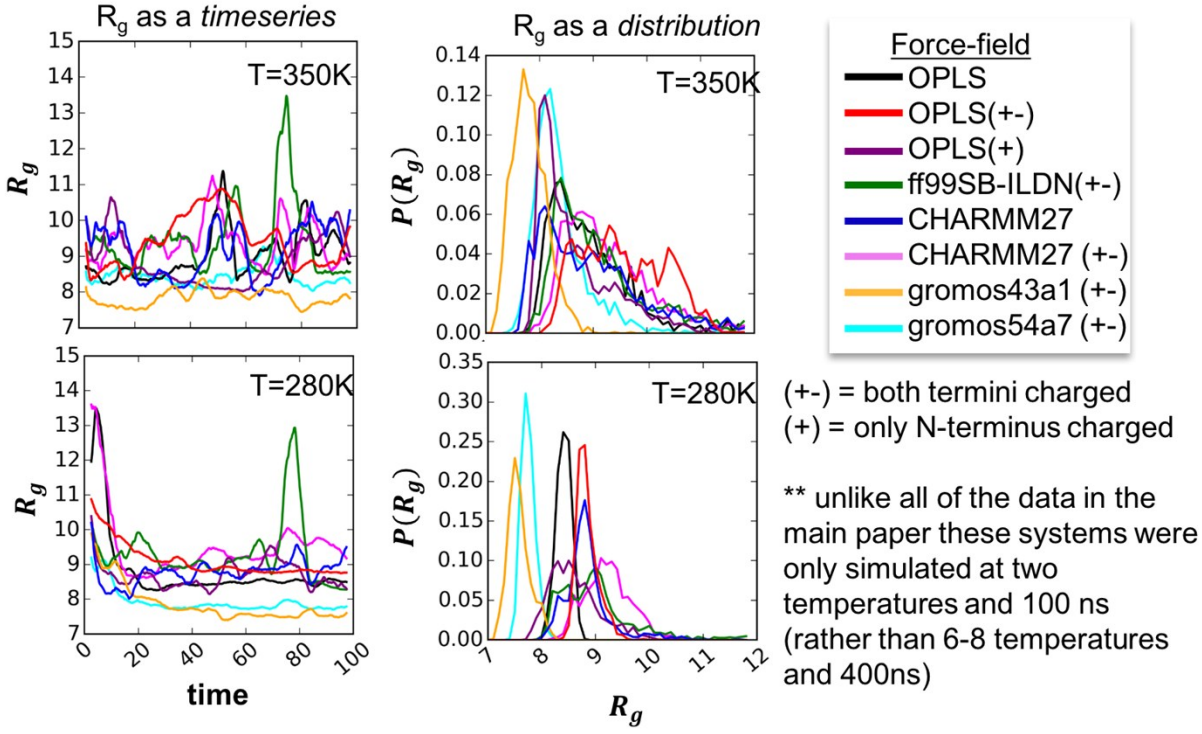


Figure S1: *Radius of gyration, R_g^2, in units of Å shown versus time in units of ns and as a probability distribution for 1 untethered ELP chain of sequence $(VPGFG)_6$ at 0.0M NaCl. The force field used for each simulation is indicated by the color as shown in the legend. For each force field, the GROMACS recommended water model was chosen.*

To assess the effect of force-field choice on the chain dimensions of an atomistically detailed ELP peptide, we ran several, short 100ns simulations at 0.0M salt. The primary result from this analysis is that while the choice of force-field does have an effect on the value of the chain dimensions, it does not seem to be affect the whether or not the inverse temperature transition is apparent. The effect of the force field seems mostly limited to whether it is an all-atom or united-atom force field (e.g. OPLS vs. GROMOS) and the charged state of the termini. Another conclusion from this analysis is that 100ns is too short of a time window to properly sample the distribution of states in these systems, as there are several long-timescale (i.e. 25-50ns) fluctuations in these data. While it would have been more instructive to repeat this analysis at a finite salt concentration, our goal with the atomistic simulations was to guide the coarse-grained simulation model, and not to conduct an exhaustive survey of the perfect atomistic force field for the system.

II. Section S2. Additional Details of Coarse-Grained Simulations

Table S1*: Simulation details for the CG simulations. All LCST polymers in this table were 20 beads long, and all rod-like blocks were 14 beads long (see Figure 2 of the main paper for the model)*

System	Cubic simulation box length (in units of σ as defined in main paper)	Number of LCST polymer chains in the system	Number of rod-like blocks
Free LCST polymers	100	9	0
3 LCST polymer chains conjugated to rod-like block	100	9	3
1 LCST polymer chains conjugated to rod-like block	100	9	9

Table S2*: Simulation protocol: Steps in the annealing schedule.*

Step	Timesteps (in millions)	Temperature (T*=kT/ε)
1	2	4.0
2	2	2.0
3	2	1.5
4	3	1.25
5	3	1.1
6	4	1.01
7	4	1.0

During annealing step 1, as shown in Table S2, each simulation is initialized at $T^* = 4.0$ for 2 million timesteps before proceeding to step 2. At step 2, the temperature of the simulation is changed instantaneously to $T^* = 2.0$ and the simulation is then run for 2 million timesteps. This process proceeds numerically through annealing steps 3 through 7 with the corresponding temperatures and timesteps for each step listed in Table S2.

Table S3: *Simulation details for the CG simulations at the same concentration but at higher number of polymer chains than in the previous CG simulations described in Table S1. All LCST polymers in this table were 20 beads long, and all rod-like blocks were 14 beads long (see Figure 2 of the main paper for the model)*

System	**Cubic simulation box length (in units of σ)**	**Number of LCST polymer chains in the system**	**Number of rod-like blocks**
Free LCST polymers	208	81	0
3 LCST polymers chains conjugated to one rod-like block	208	81	27

III. Section S3. Additional Results

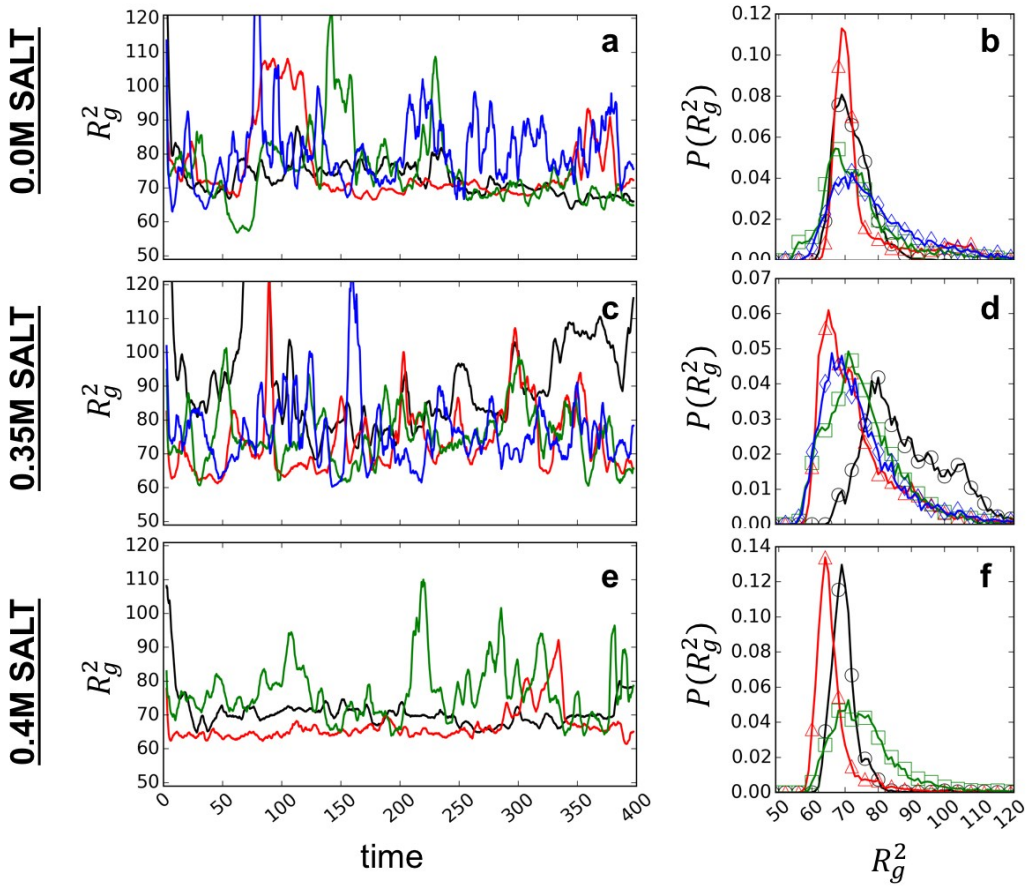


Figure S2: *(a,c,e) Squared radius of gyration, R_g^2, in units of Å, shown versus time, in units of ns, and (b,d,f) as a probability distribution for 1 untethered ELP chain of sequence $(VPGFG)_6$ at 0.0M (a,b) 0.35M (c,d) and 0.4M (e,f) NaCl. The colors and symbols correspond to the simulation temperature: 280K (circles), 320K (triangles), 360K (squares), 400K (diamonds). The data in parts a,c,e was smoothed using window averaging with a width of 5 ns.*

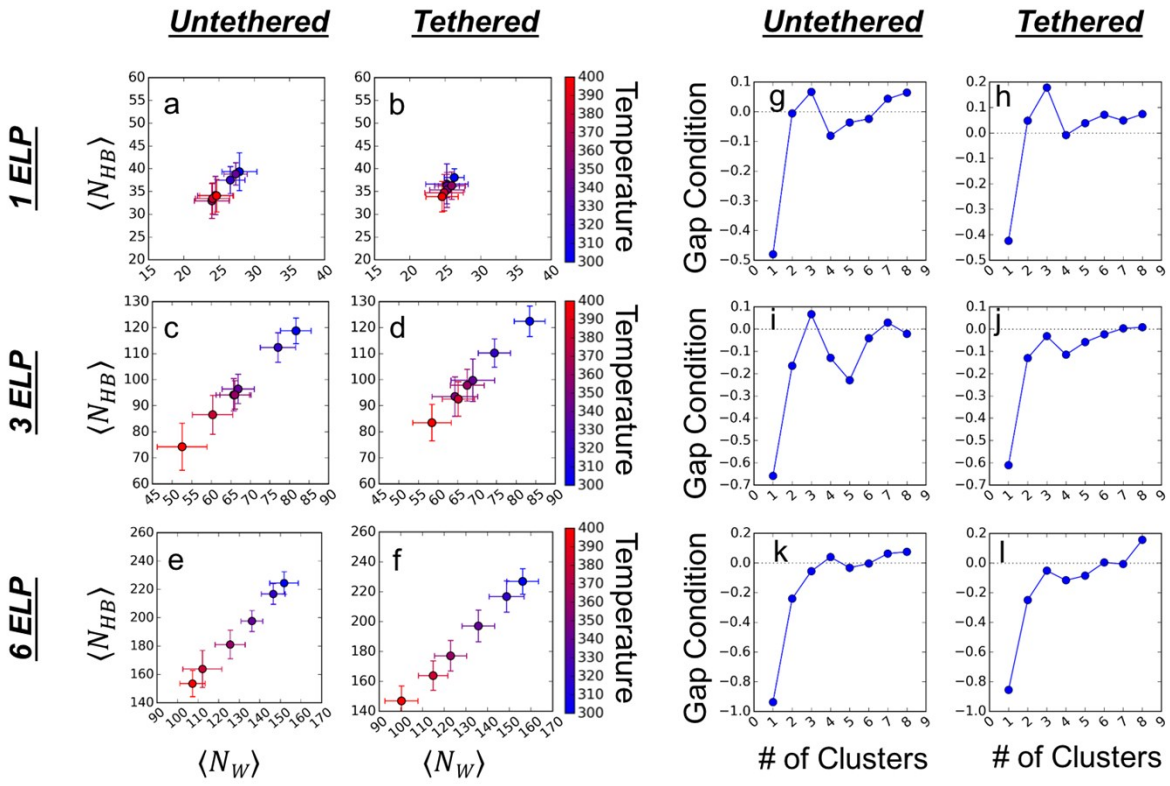


Figure S3: *(a-f) Average number of hydrogen peptide-water bonds,* $\langle N_{HB} \rangle$*, versus the average number of water molecules within 2.43* Å *of the peptide backbone* $\langle N_W \rangle$*, with the simulation temperature as indicated by the colorbar. (g-l) Gap statistic condition versus the number of data clusters input into the K-means algorithm. All data for parts a-e is for the ELP chain of sequence* $(VPGFG)_6$ *at 0.3M NaCl. The number of peptides and their (un)tethered state are indicated by the row and column headers, respectively.*

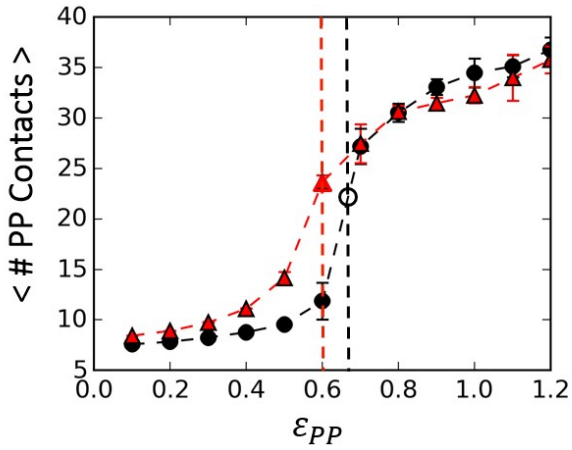
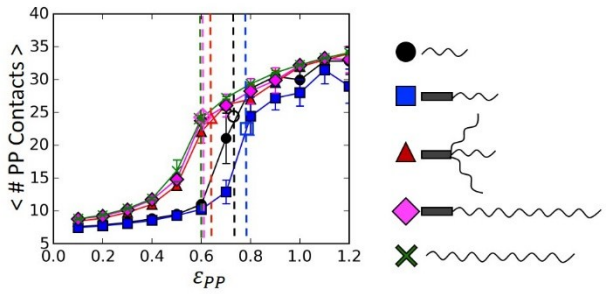


***Figure S4**: **Larger system sizes simulations.** Average number of polymer-polymer contacts vs. ε_{PP}. The closed black circles represent the free LCST polymer system with 81 polymer chains, 20 polymer (P) beads per polymer chain. The closed red triangles represent the diblock system with 27 diblocks where each rod-like block is 14 rod (C) beads long, with three 20mer polymer chains conjugated to each rod-like block. The red and black vertical dashed lines for the two systems denote the onset of aggregation.*

***Table S4**: Aggregation transitions for varying the number of LCST polymers grafted to rod-like blocks, for larger systems with 81 polymer chains per simulation box.*

System	ε_{Agg} (logistic fit)	ε_{Agg} (linear interpolation)
81 free polymers	0.679	0.673
27 diblocks. Each diblock has 3 polymer chains conjugated to a rod-like block	0.601	0.588



System	ε_{Agg} (logistic fit)	ε_{Agg} (linear interpolation)
9 20mer LCST polymers	0.706	0.709
9 diblocks with 1 20mer LCST polymer chains per diblock	0.760	0.766
3 diblocks with 3 20mer LCST polymer chains per diblock	0.612	0.594
3 diblocks with 1 60mer LCST polymer chains per diblock	0.593	0.577
3 60mer LCST polymers	0.581	0.579

Figure S5*: Average number of polymer-polymer contacts vs. ε_{PP}. The legend and the table (first column) describe the systems presented in the plot. The vertical dashed lines denote the onset of aggregation also listed in two columns in the table.*

In Figure S5 we provide additional proof that increasing polymer bead crowding leads to decreasing loss in translational entropy of the polymer beads upon aggregation $T\Delta S_{Agg}$, and as a result, a lower energetic driving force (ε_{Agg}) for aggregation. One should note that all of these systems in Figure S5 have the same number of total polymer beads and thus, same total *possible* PP contacts. First, we compare the system with three 20mer tethered LCST chains (red triangles) to the system with single 60mer tethered LCST chains (pink diamonds). Our simulations show that these two systems have a similar onset of aggregation. In both cases, the 60 LCST polymer (P) beads are crowded near the rod-like block (i.e. number of P beads effectively attached to each rod-like block in the system is the same). Furthermore, the tethering effect is similar, whether the three 20mer blocks are all tethered to a rod-like block (red triangles case) or sequentially tethered to a rod-like block and then each other (pink diamonds case). In both cases, the translation entropy of the monomers is restricted by being bound to one another and to the rod-like block. Next, we compare the system with three diblocks with one 60mer tethered LCST chain (pink diamonds in Figure S5) and the system with three 60mer free LCST chains (crosses in Figure S5). Surprisingly, these two systems do not show a difference in ε_{Agg} while the analogous 20mer systems (blue squares and black circles) show a difference. We conjecture that the effect of the rod-block on the ε_{Agg} diminishes as the polymer chain length increases (while the rod block length remains constant) because the large number of polymer beads dominate the systems' $T\Delta S_{Agg}$ and the energetic driving force needed to balance that loss upon aggregation.

Table S5: *Aggregation transitions for varying flexibility of second block to which the LCST polymer is conjugated*

System	**ε_{Agg} (logistic fit)**	**ε_{Agg} (linear interpolation)**
9 free polymers	0.706	0.709
9 diblocks. Each diblock has 1 polymer attached to a rigid block	0.760	0.765
9 diblocks. Each diblock has 1 polymer attached to a flexible block	0.753	0.752