Supplemental Material:

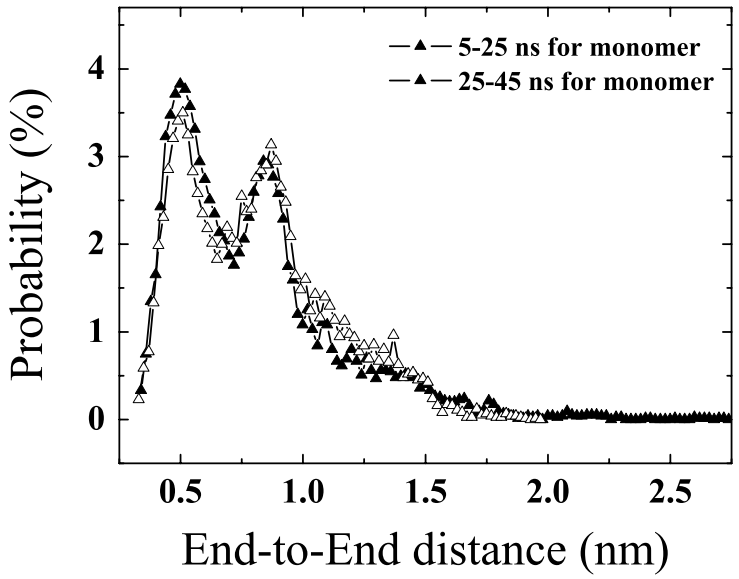

Figure S1: Testing the convergence of the REMD monomer simulation. This figure compares the distribution of peptide C_α end-to-end distances at 310 K, using the first and last 20 ns of simulation data. (The first 5 ns were discarded.) The agreement between the two distributions suggests the monomer simulation has equilibrated.

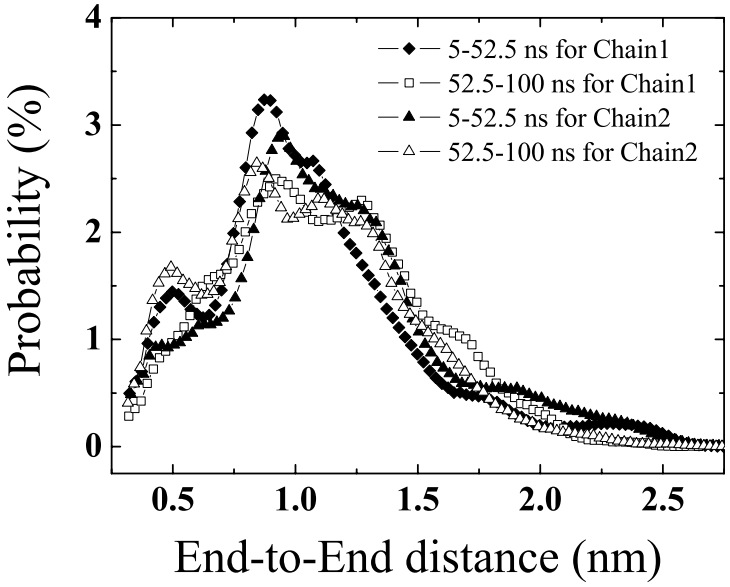

Figure S2: Testing the convergence of the REMD dimer simulation. This figure compares the distribution C_α end-to-end distances at 310 K, using first and last 47.5 ns of simulation data. (The first 5 ns were discarded.) The distributions corresponding to different chains and different time intervals are all in reasonable agreement.

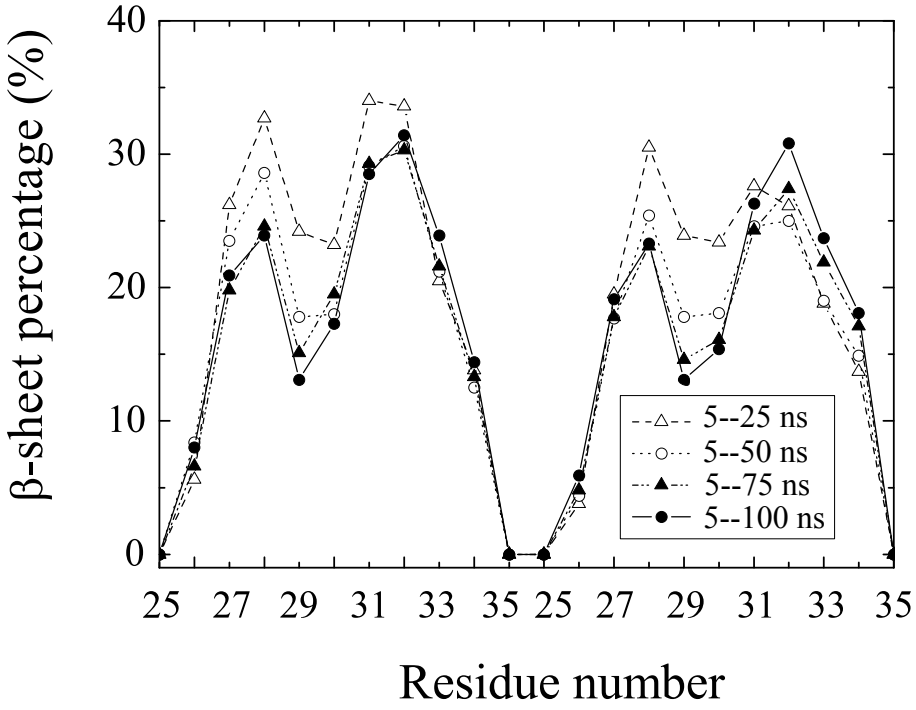

Figure S3: Testing the convergence of the REMD dimer simulation by examining the time evolution of the average secondary structure. Here we calculate the percentage of time that each residue spent belonging to a β-sheet during the following time intervals: 5-25 ns, 5-50 ns, 5-75 ns and, 5-100 ns. (The secondary structure of each amino acid in both peptides was determined using the DSSP program, as explained in the Materials and Methods.) The difference between the data, once averaged over 5-75 ns, compared to 5-100 ns has decayed to less than 4%, suggesting that by 75 ns, the REMD simulation has converged.

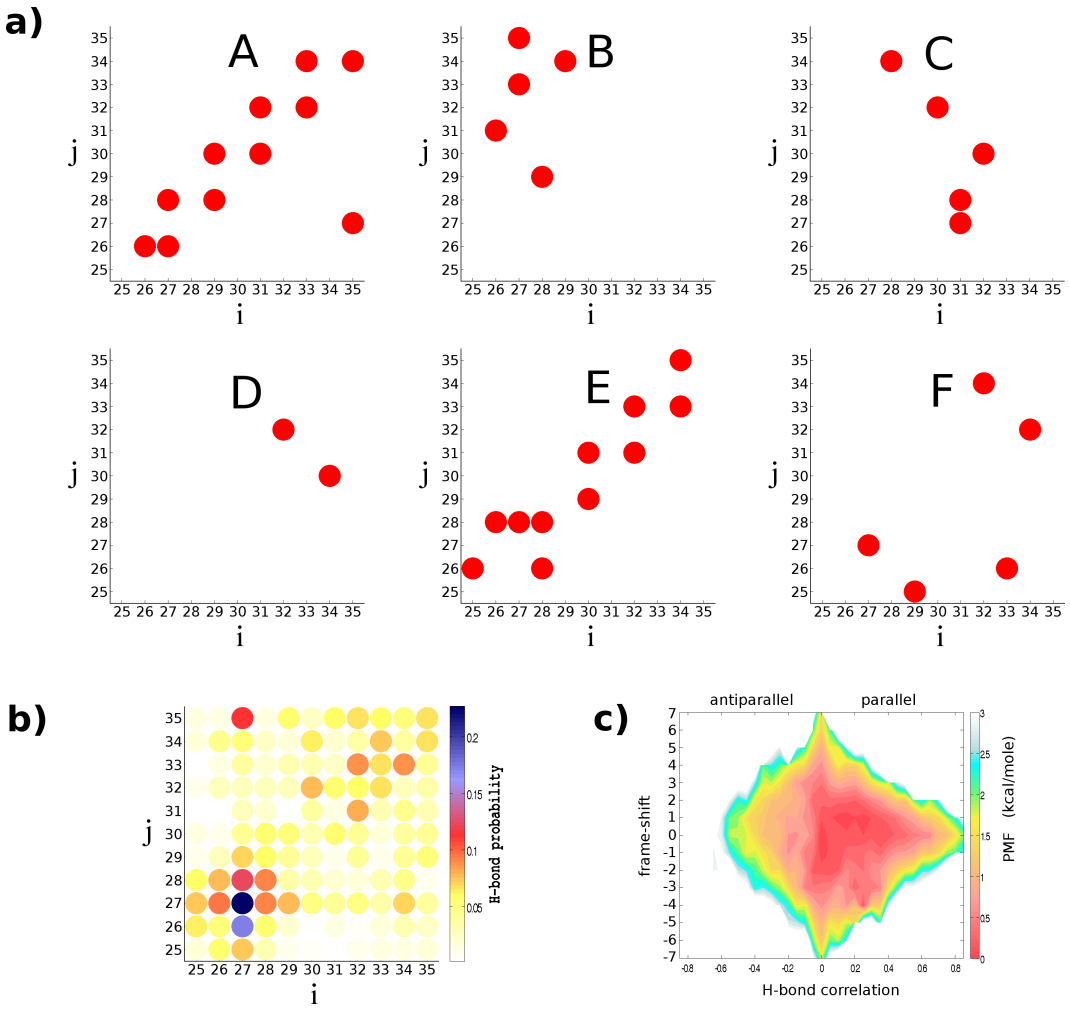

Figure S4: Inter-peptide hydrogen-bonding patterns are shown **a)** for each of the cluster centers shown figure 4a), and **b)** averaged over all the structures visited at 300 K. In part **a)**, a red dot indicates that one (or more) hydrogen bonds exists connecting amino acid i from peptide 1, with amino acid j from peptide 2 in this structure (A,B,C,D,E, or F). The overall probability that amino acid i from peptide 1 has a hydrogen-bond with j from peptide 2, is shown in part **b)**. This probability is computed from all of the conformations sampled at 310 K (after discarding the first 10 ns). **c)** Here we show the probability at 300 K that the dimer is parallel or antiparallel, as well as the probability that peptides are in-register or out-of register. This information is expressed as a potential of mean force (PMF). The vertical axis indicates the frame shift for a given conformation, which is defined as: $\langle j \rangle - \langle i \rangle$, where $\langle i \rangle = \frac{1}{N} \sum_{n=1}^{N} i_n$, (and i_n and j_n are integers ranging from $25 \ldots 35$ indicating the location in chain 1 and chain 2 of the n'th inter-peptide hydrogen bond, and N is the number of inter-peptide hydrogen bonds for a particular conformation.) The horizontal axis, "H-bond correlation" for a given conformation is defined as the Pearson's correlation between the amino acids participating in hydrogen bonds between the two peptides. (We estimate this using: $\frac{1}{N-1} \sum_{n=1}^{N} (i_n - \langle i \rangle)(j_n - \langle j \rangle)$ / δ_i δ_j), and $\delta_i = \sqrt{\frac{1}{N-1} \sum_{n=1}^{N} (i_n - \langle i \rangle)^2}$).