

The Supporting Information contain a comparison of the current GXYG analysis with the one of Toal et al., Tables listing the parameters used for the new analysis (Tables S1-S3), configurational entropy values for GXYG (Table S4), a truncated Ramachandran plot scheme used for the analysis of amide I' profiles (Figure S1), a drawing illustrating the mesostates (Figure S2), bar plots depicting statistical weights obtained from the new Gaussian analysis of GXYG (Figures S3-S7), plots of the Gibbs free energy difference between pII and  $\beta$ -strand conformations for different total fractions of the two conformations (Figure S8), Ramachandran plots of GXG, GXYG, all-XG, all-XY, GY-all and XY-all peptides/coli library dimers (Figure S9-S16), mesostate populations of individual residues in GXYG peptides and corresponding XY-coil library dimers (Figure S17), a comparison of  $^3J(\text{H}^{\text{N}}\text{H}^{\text{C}\alpha})$  values of GXYG and blocked dipeptides with two residues (Figure S18) and a two-dimensional probability density plot of serine in all-SL as a function of  $\phi$  (Figure S19).