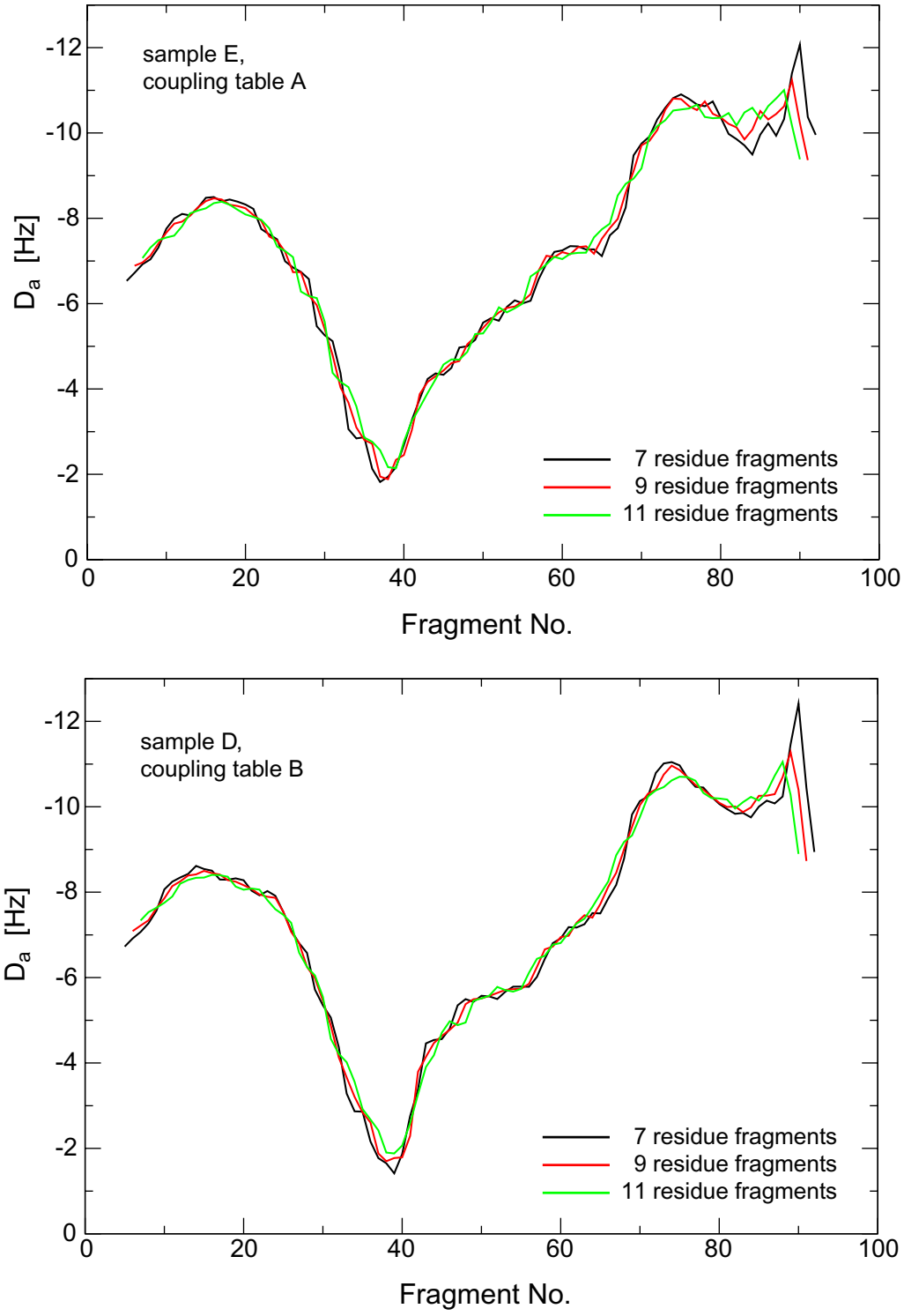
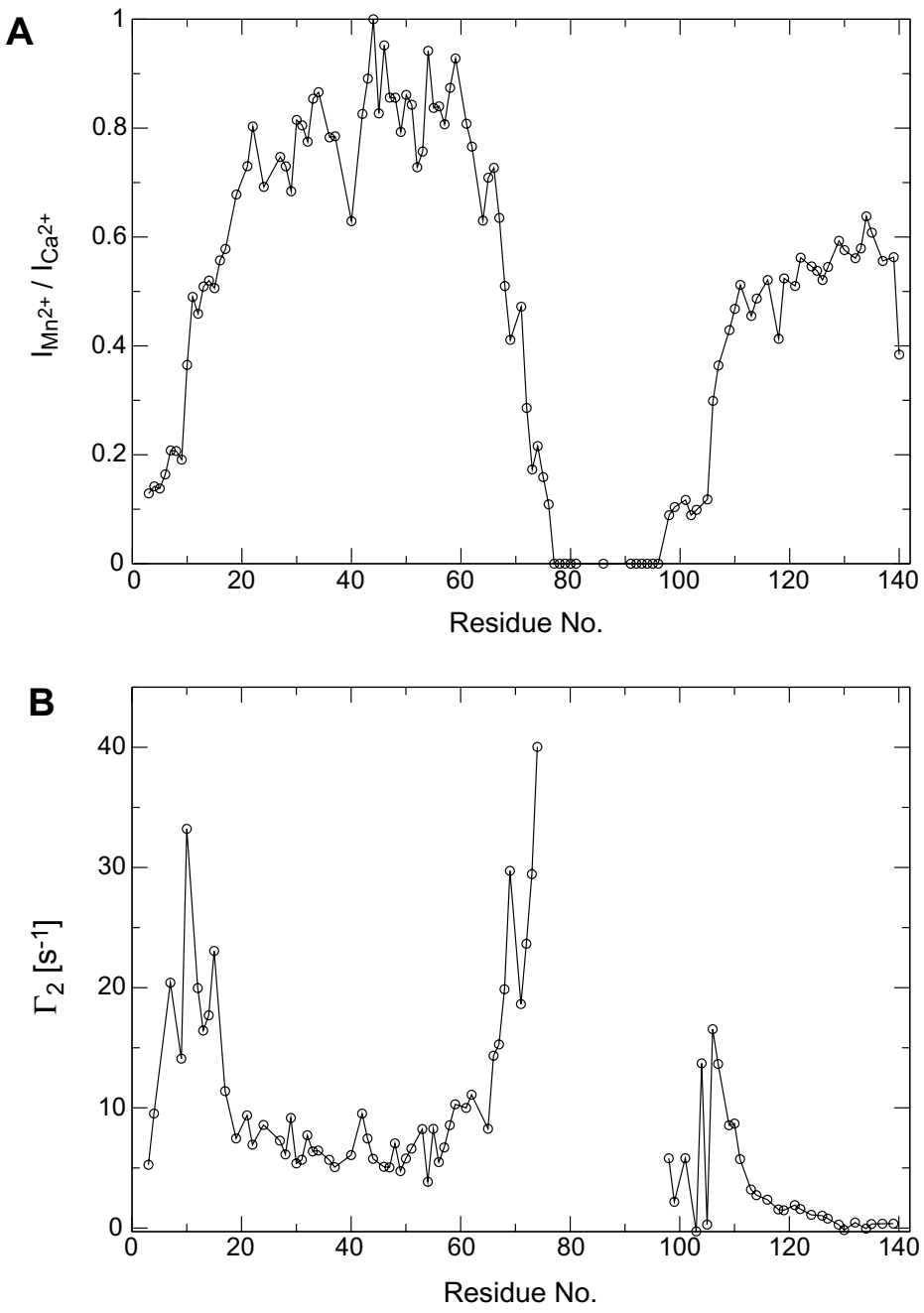
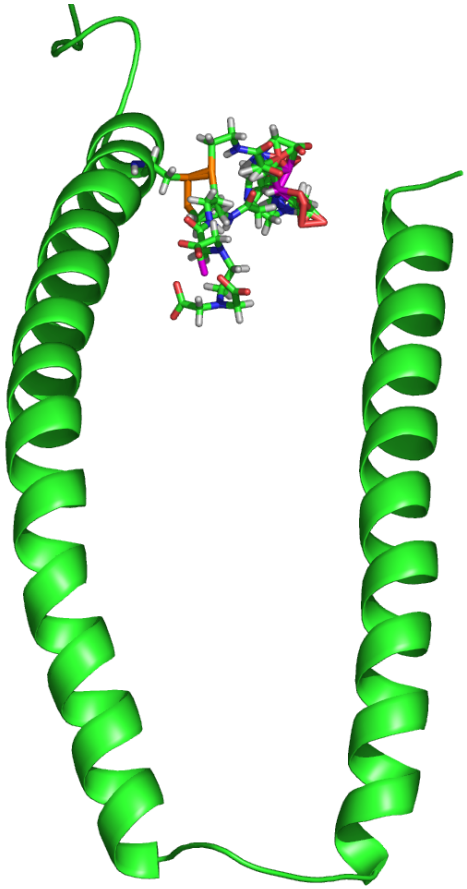


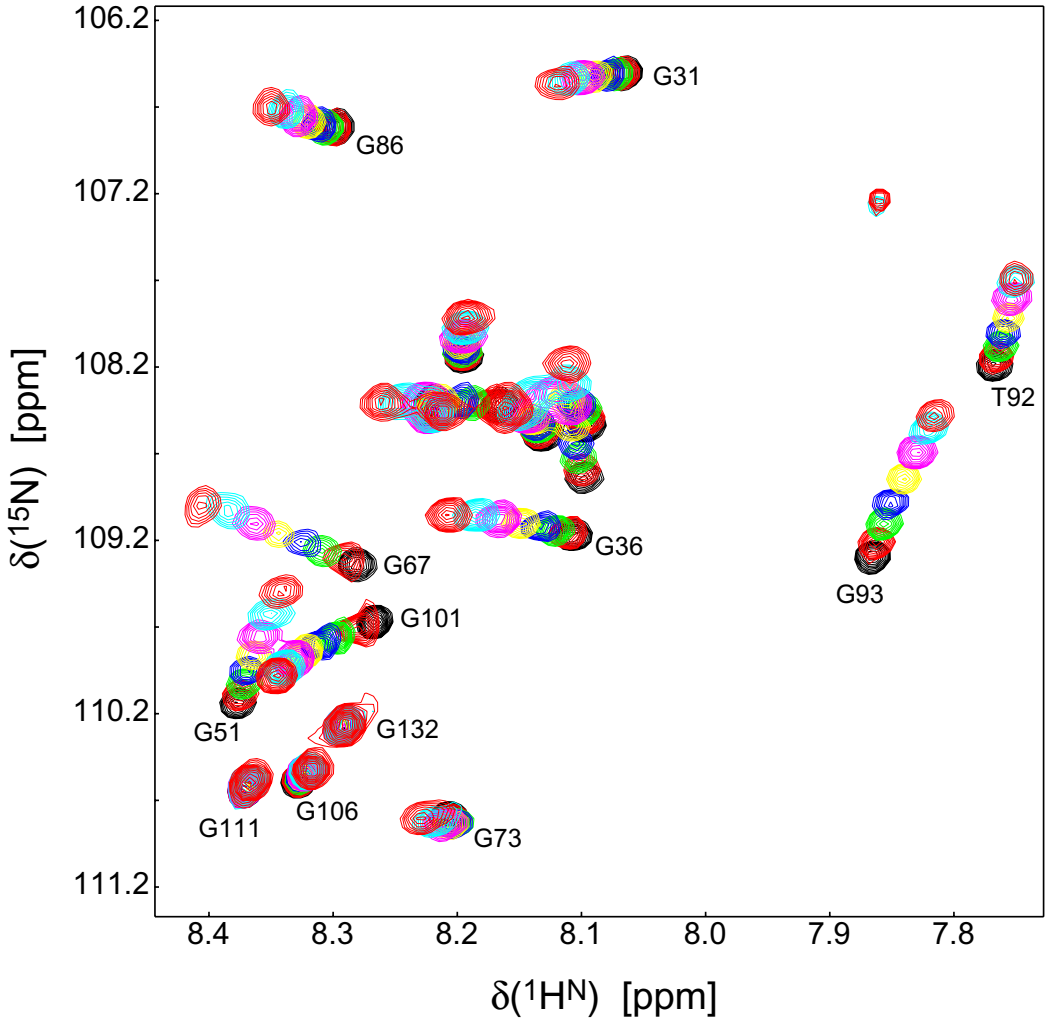
Supplementary Figure 1. Variation of Da with fragment length. The fragment number is the center residue number of each fragment.



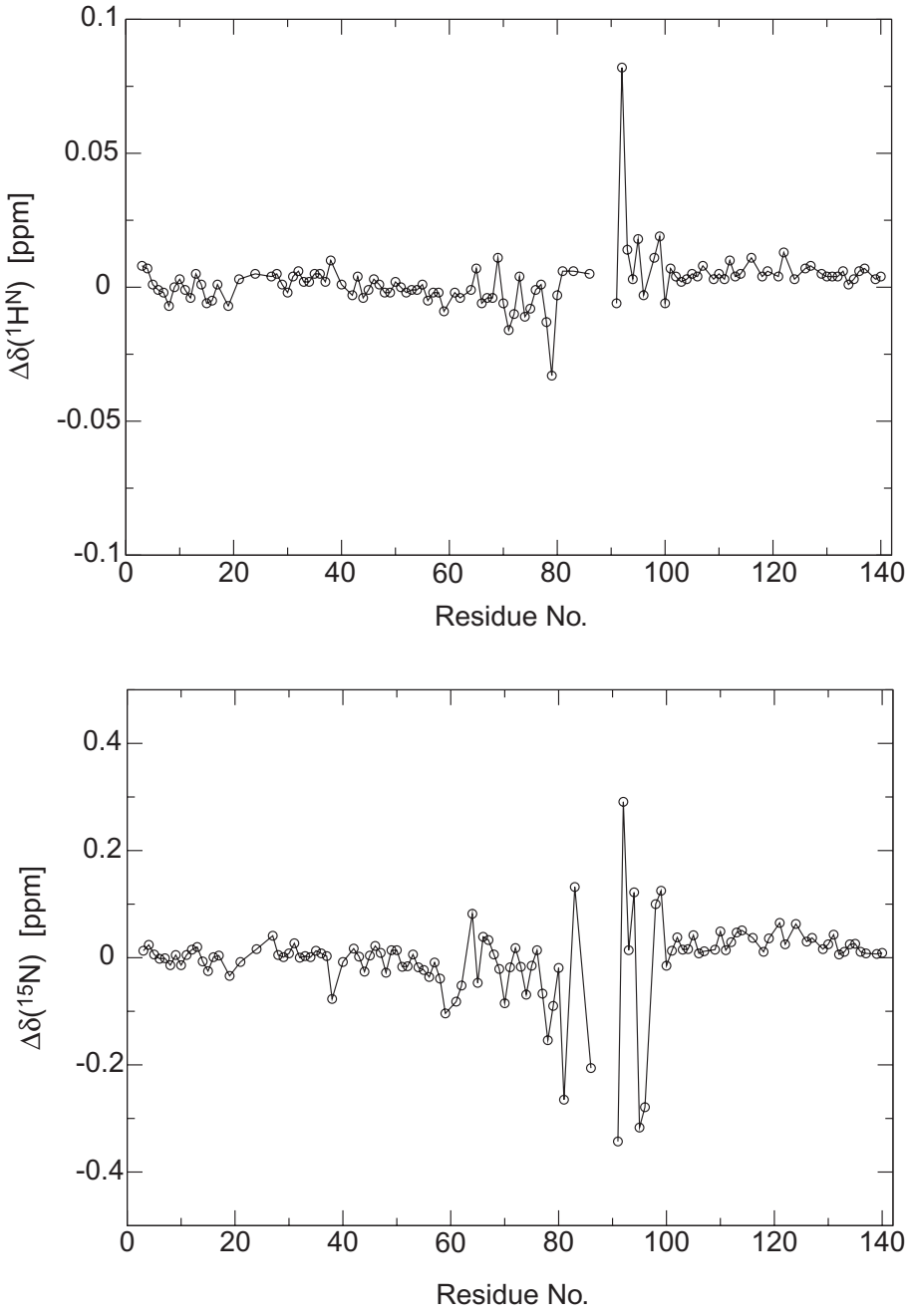
Supplementary Figure 2. (**A**) Intensity ratio of TROSY-HSQC H-N resonances of aS(S87C)-cysteaminyl-EDTA complexed with Mn^{2+} and Ca^{2+} (samples F and G), respectively, at 25 °C and 800 MHz. (**B**) Mn^{2+}-induced paramagnetic relaxation enhancements, Γ_2, of H^N nuclei at 25 °C and 600 MHz.



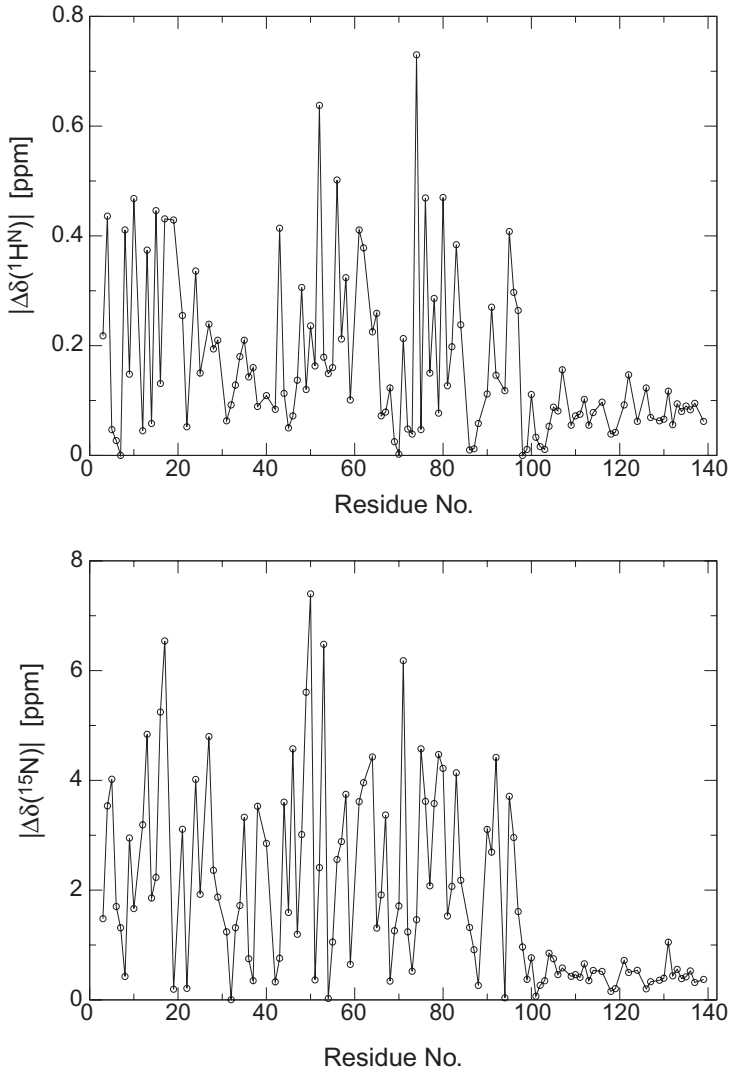
Supplementary Figure 3. Ensemble representative of aS showing the three cysteaminyl-EDTA-Mn^{2+} tags, representing the conformational space accessible to Mn^{2+}, attached to Cys87. All tags are cleary orientied to one side of helix-C, indicating the relative orientation of helix-N and -C.



Supplementary Figure 4. Superimposed region of 2D H-N correlation spectra of aS. A titration has been performed between molar SDS:DPC ratios of 100:0 to 30:70 in eight steps, allowing the transfer of assignments between both states. The spectra in 100:0 SDS:DPC is shown in black; during the titration the ratio is changed in steps of 10%. Spectra were recorded at 25 °C and 750 MHz in 20 mM NaH2PO4/Na2HPO4, pH 7.4, with starting concentrations of [aS]=0.2 mM and [SDS]=30 mM.



Supplementary Figure 5. Chemical shift differences between w.t. aS and aS(S87C), samples B and F, respectively (c.f. Methods).



Supplementary Figure 6. Absolute chemical shift differences between micelle-bound aS and aS in the absence of SDS micelles. Chemical shifts in the absence of SDS micelles courtesy of Markus Zweckstetter.