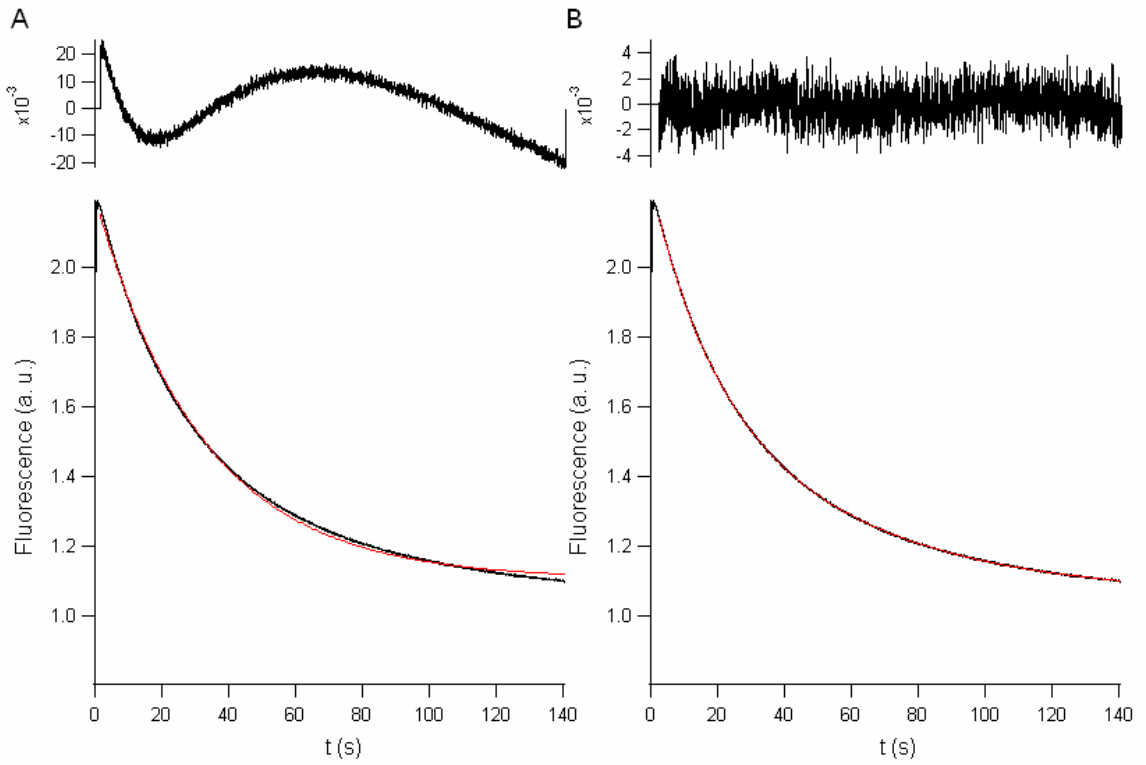

Figure 1S. A comparison between single exponential fit (A) and double exponential fit (B) to the fluorescence decay phase of GL5/I27w34f in 0.3M GdmCl. Red lines are fits to the experimental data, and top panels are plots of residuals from the fitting. It is evident that the fluorescence decay phase is best described by a double exponential fit. However, the underlying molecular mechanism for such a double exponential folding behavior remains unclear.

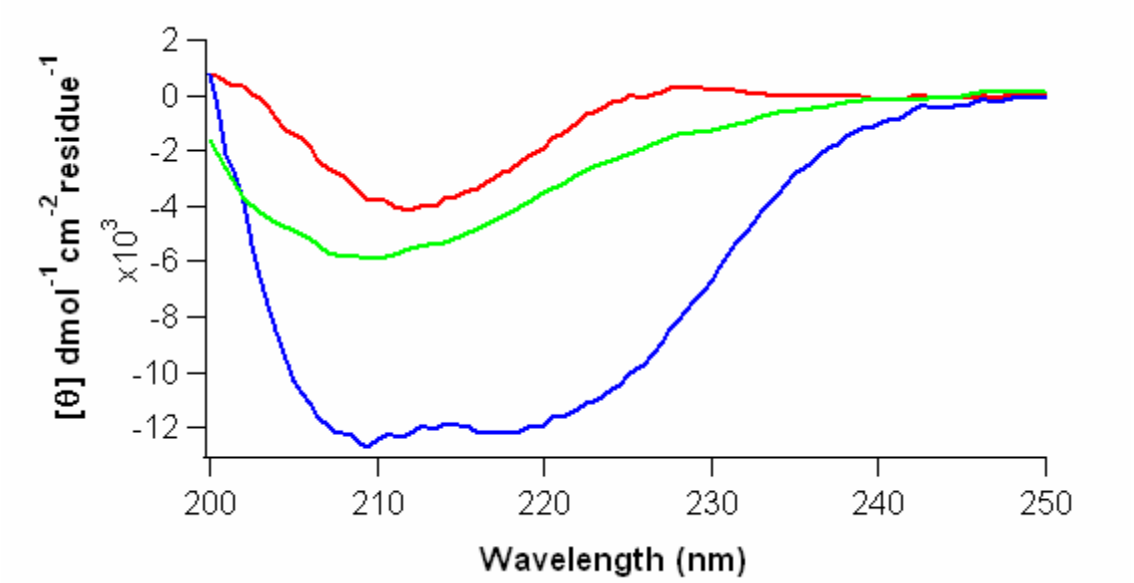

Figure 2S. Far-UV CD Spectra of isolated GL5 (blue line), isolated I27w34f (red line) and combined GL5/I27w34f (green line) in PBS buffer at pH 7.4. The CD spectrum of GL5/I27w34f clearly indicates the disappearance of the majority of α-helical structures, suggesting that the host GL5 domain is largely unfolded. This is in good agreement with the conclusion that the majority of mutually exclusive protein GL5/I27w34f exists in the form of $GL5_{unfolded}$/$I27w34f_{folded}$ in PBS buffer and in solutions containing low concentration of GdmCl.

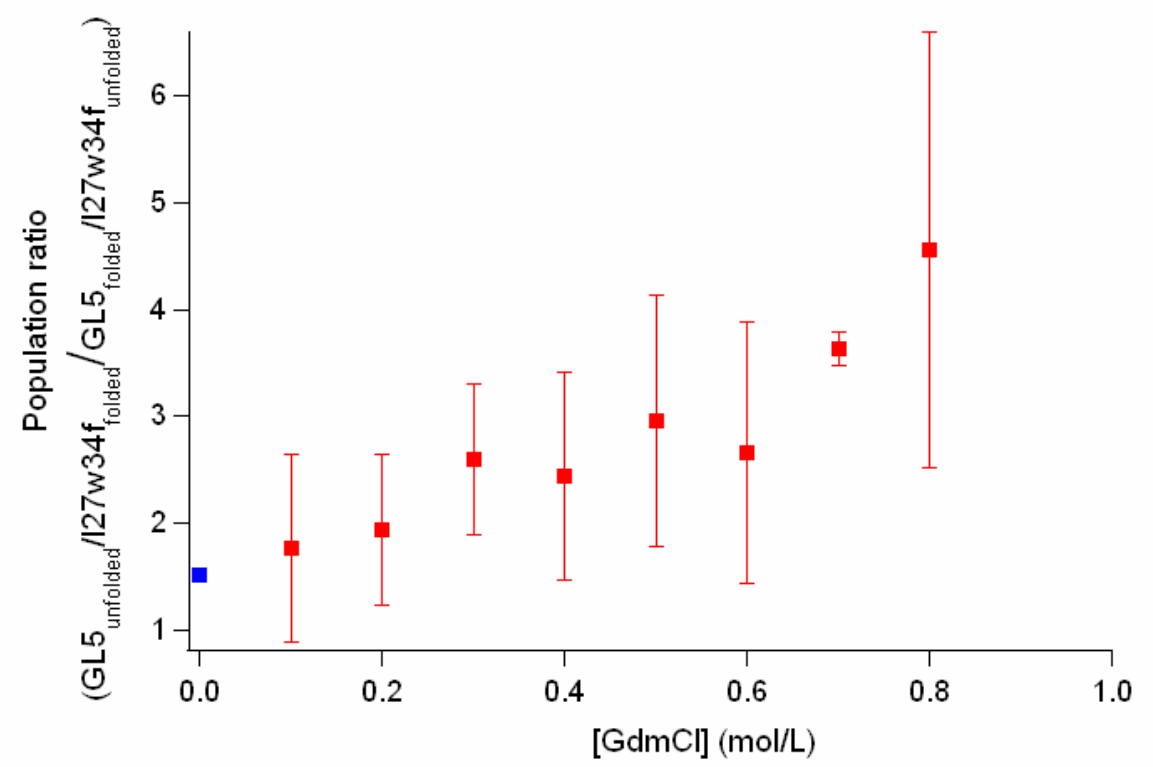

Figure 3S. Conformational equilibrium between $GL5_{folded}$/I27w34f $_{unfolded}$ and $GL5_{unfolded}$/I27w34f $_{folded}$ depends on the concentration of GdmCl. Red symbols are the ratio between $GL5_{folded}$/$I27w34f_{unfolded}$ and $GL5_{unfolded}$/$I27w34f_{folded}$ measured from fluorescence decay experiments, and the blue symbol is measured from single molecule AFM experiments. It is evident that the ratio measured in AFM is in reasonable agreement with that measured from fluorescence decay experiments.