

Supporting Information

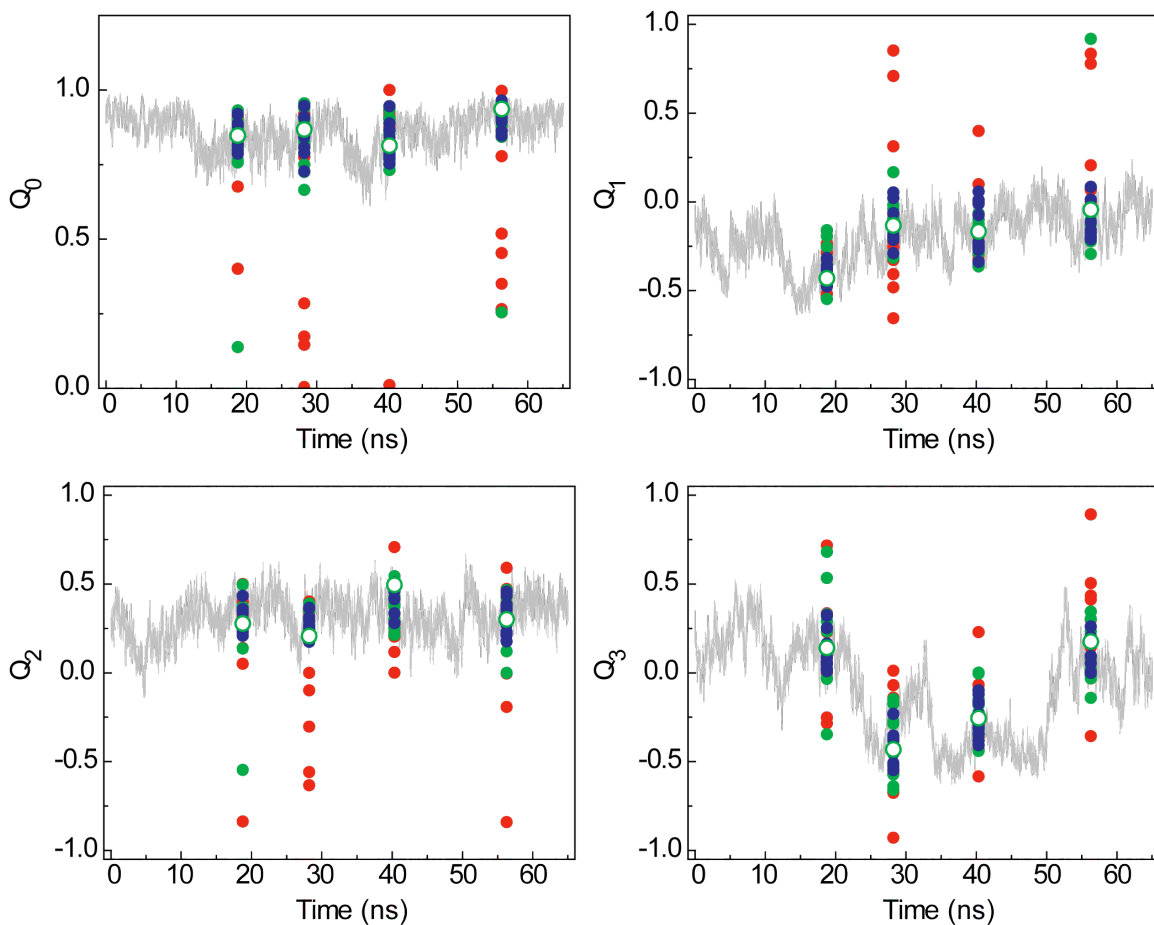


Figure S1. 4-state ensemble fit to $\langle D_{nk}^2(\alpha\beta\gamma) \rangle_{real}$ computed for a motional model involving collective motions of two helical domains in a 65 ns molecular dynamics simulation of TAR RNA^{25,34}. Comparison of the MD trajectory (shown in gray) and 4 (open green circles) state ensemble. Also shown are ensembles calculated for 10 iterations with three levels of error; (blue circles) $\leq 5^\circ$ uncertainty in orientation and $\leq 5\%$ uncertainty in magnitude, (green circles) 5° - 10° uncertainty in orientation and 5-10 % uncertainty in magnitude, (red circles) 10° - 15° uncertainty in orientation and 10-15 % uncertainty in magnitude. The order of the four states was chosen to minimize the overall RMSD compared to the zero error ensemble. Each state defined as an inter-helical

domain orientation is represented using a unit quaternion; a 4-dimensional complex number that represents an orientation as a single axis rotation from the z-axis. The relationship between the quaternions and the Euler angles is given in Equation 14. Shown are the values for the four quaternion components; Q_0 , Q_1 , Q_2 , and Q_3 . Q_0 encodes the rotational amplitude and Q_1 , Q_2 , and Q_3 encode the x, y and z components of the rotation axis, respectively.

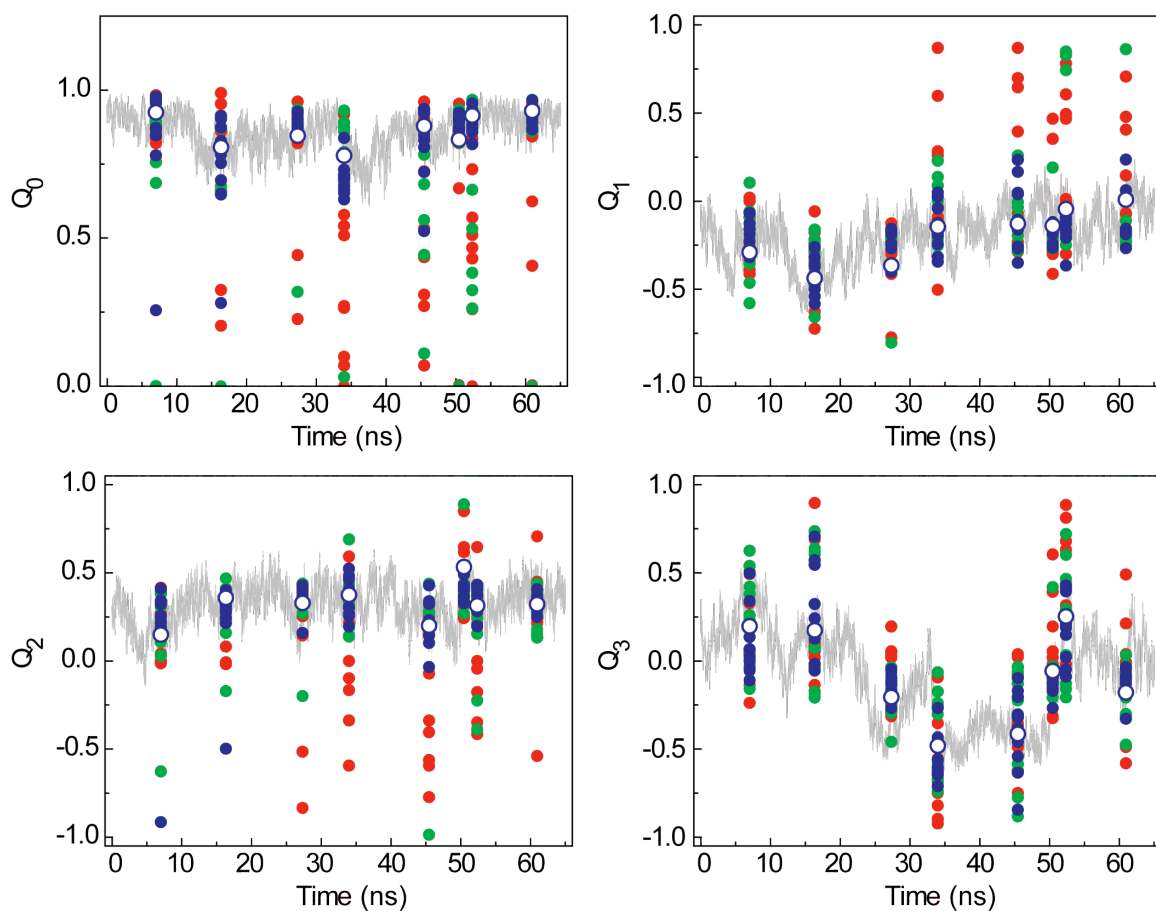


Figure S2. 8-state ensemble fit to $\langle D_{nk}^2(\alpha\beta\gamma) \rangle_{real}$ computed for a motional model involving collective motions of two helical domains in a 65 ns molecular dynamics simulation of TAR RNA^{25,34}. Comparison of the MD trajectory (shown in gray) and 8

(open blue circles) state ensemble. Also shown are ensembles calculated for 10 iterations with three levels of error; (blue circles) $\leq 5^\circ$ uncertainty in orientation and $\leq 5\%$ uncertainty in magnitude, (green circles) 5° - 10° uncertainty in orientation and 5 - 10% uncertainty in magnitude, (red circles) 10° - 15° uncertainty in orientation and 10 - 15% uncertainty in magnitude. The order of the eight states was chosen to minimize the overall RMSD compared to the zero error ensemble. Each state defined as an inter-helical domain orientation is represented using a unit quaternion; a 4-dimensional complex number that represents an orientation as a single axis rotation from the z-axis. The relationship between the quaternions and the Euler angles is given in Equation 14. Shown are the values for the four quaternion components; Q_0 , Q_1 , Q_2 , and Q_3 . Q_0 encodes the rotational amplitude and Q_1 , Q_2 , and Q_3 encode the x, y and z components of the rotation axis, respectively.