Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information: Conformational Properties of Modified Nucleobases in RNA Aptamers and their Effect on Förster Resonant Energy Transfer

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Molecular Dynamics Parameters

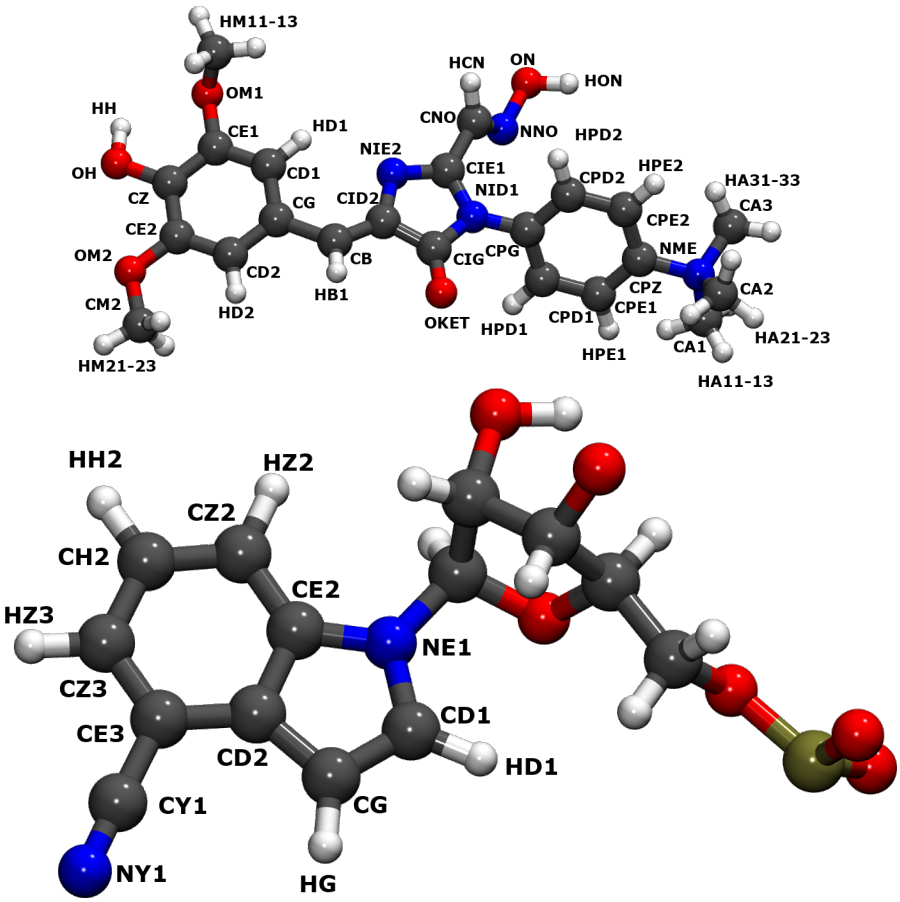

Figure 1: Depiction of the atom name assignment of DMHBO$^+$ (top) and r4CI (bottom) for the forcefield parameters.

The parameters used for DMHBO$^+$ and r4CI we utilized are as follows:

DMHBO$^+$ and r4CI CHARMM36 Parameters

```
! r4CI (CYRI) residue
RESI CYRI          -1.00 ! CYRI C8H7N indole, adm jr., atm
ATOM P     P        1.50
ATOM O1P   ON3     -0.78
ATOM O2P   ON3     -0.78
ATOM O5'   ON2     -0.57
ATOM C5'   CN8B    -0.08
ATOM H5'   HN8      0.09
ATOM H5''  HN8      0.09
GROUP
ATOM C4'   CN7      0.16
ATOM H4'   HN7      0.09
ATOM O4'   ON6B    -0.50
ATOM C1'   CN7B     0.16
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```
ATOM H1'   HN7          0.09
ATOM C3'   CN7          0.01
ATOM H3'   HN7          0.09
ATOM O3'   ON2         -0.57
ATOM C2'   CN7B         0.14
ATOM H2''  HN7          0.09
ATOM O2'   ON5         -0.66
ATOM H2'   HN5          0.43
GROUP
ATOM  HG       HGR51    0.1641   !   0.14 !
ATOM  CG       CG2R51   -0.3119  !  -0.17 !
ATOM  CD2      CG2RC0   0.4834   !   0.11 !
ATOM  CD1      CG2R51   -0.7254  !  -0.15 !
ATOM  HD1      HGR52    0.5450   !   0.22 !
ATOM  NE1      NG2R51   0.8213   !  -0.51 !
!TOM HE1   HGP1       123   !   0.37 !
ATOM  CE2      CG2RC0   -0.7012  !   0.24 !
ATOM  CE3      CG2R61   -0.2905  !  -0.25 !
!TOM HE3   HGR61      123   !   0.17 !
ATOM  CZ2      CG2R61   0.0012   !  -0.27 !
ATOM  HZ2      HGR61    0.1117   !   0.16
ATOM  CZ3      CG2R61   0.0119   !  -0.20
ATOM  HZ3      HGR61    0.0910   !   0.14
ATOM  CH2      CG2R61   -0.1920  !  -0.14
ATOM  HH2      HGR61    0.0875   !   0.14
ATOM  CY1      CG1N1    0.4464   !   0.39
ATOM  NY1      NG1T1    -0.5426  !  -0.47
BOND P    O1P
BOND P    O2P
BOND P    O5'
BOND C5' O5'
BOND C5' H5'
BOND C5' H5''
BOND C5' C4'
BOND C4' H4'
BOND C4' O4'
BOND C4' C3'
BOND C3' O3'
BOND C3' H3'
BOND C3' C2'
BOND C2' C1'
BOND C2' H2''
BOND C2' O2'
BOND O2' H2'
BOND C1' H1'
BOND C1' O4'
BOND C1' NE1
BOND NY1 CY1
BOND CY1 CE3
BOND CG HG CD2 CG NE1 CD1
BOND CZ2 CE2
BOND CZ3 CH2 CD2 CE3 NE1 CE2
BOND CD1 HD1
BOND CZ3 HZ3 CH2 HH2 CZ2 HZ2
DOUBLE CD1 CG CE2 CD2 CH2 CZ2 CZ3 CE3
IC CG    CD1   NE1   CE2    1.3650   110.50     0.00   112.00    1.3700
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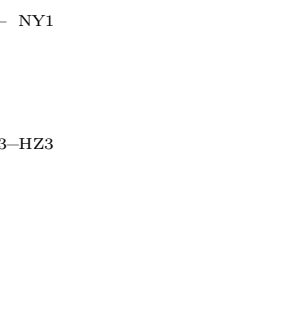

```
IC CD1  CG   CD2  CE2   1.3650  106.40    0.00  108.00   1.3850
IC CD2  CG   CD1  NE1   1.4300  106.40    0.00  110.50   1.3700
IC CE2  CG   *CD2 CE3   1.3850  108.00  180.00  133.50   1.3600
IC CE2  CD2  CE3  CZ3   1.3850  110.00    0.00  113.20   1.3750
IC CD2  CE3  CZ3  CH2   1.3600  113.20    0.00  120.00   1.3750
IC CE3  CZ3  CH2  CZ2   1.3750  120.00    0.00  120.00   1.3750
IC CH2  CE3  *CZ3 HZ3   1.3750  120.00  180.00  120.00   1.0800
IC CZ2  CZ3  *CH2 HH2   1.3750  120.00  180.00  120.00   1.0800
IC CE2  CH2  *CZ2 HZ2   1.3600  113.20  180.00  120.00   1.0800
IC CD1  CE2  *NE1 HE1   1.3700  112.00  180.00  126.00   0.9760
IC CG   NE1  *CD1 HD1   1.3650  110.50  180.00  125.00   1.0800
IC CD1  CD2  *CG  HG    1.3650  106.40  180.00  126.40   1.0800
PATCH FIRST NONE LAST NONE

! DMHBO+ (HBO) residue
RESI HBO 1.00 ! HBO
GROUP
ATOM  CG       CG2R61  0.3153
ATOM  CD1      CG2R61  -0.6582
ATOM  HD1      HGR61   0.2757
ATOM  CD2      CG2R61  -0.6126
ATOM  HD2      HGR61   0.2420
ATOM  CE1      CG2R61  0.6210
ATOM OM1   OG301       -0.34
ATOM CM1   CG331       -0.10
ATOM HM11 HGA3          0.09
ATOM HM12 HGA3          0.09
ATOM HM13 HGA3          0.09
GROUP
ATOM  CE2      CG2R61  0.6210
ATOM OM2   OG301       -0.34
ATOM CM2   CG331       -0.10
ATOM HM21 HGA3          0.09
ATOM HM22 HGA3          0.09
ATOM HM23 HGA3          0.09
GROUP
ATOM  CZ       CG2R61  -0.1850
ATOM  OH       OG311   -0.4514
ATOM  HH       HGP1    0.3811
GROUP
ATOM  CB       CG2DC3  -0.1149
ATOM  HB1      HGA4    0.1209
GROUP ! IMIDAZOLE MEOI MRDN
ATOM  CIG      CG2R53  0.3870
ATOM  CID2     CG25C2  0.2205
ATOM  NID1     NG2R51  0.0090
ATOM  CIE1     CG2R53  0.0665
ATOM  CNO      CG2DC1  0.3871
ATOM  NIE2     NG2R50  -0.4958
ATOM  HCN      HGA4    0.0749
GROUP
ATOM  NNO      NG2D1   -0.2959
ATOM  ON       OG311   -0.4298
ATOM  HON      HGP1    0.4011
ATOM  OKET     OG2D1   -0.5494
GROUP ! PARA BENZ
```

```
ATOM   CPG       CG2R61   0.1874
ATOM   CPD1      CG2R61   -0.1126
ATOM   HPD1      HGR61    0.1751
ATOM   CPD2      CG2R61   -0.3046
ATOM   HPD2      HGR61    0.2406
ATOM   CPE1      CG2R61   -0.4905
ATOM   HPE1      HGR61    0.2234
ATOM   CPE2      CG2R61   -0.2023
ATOM   HPE2      HGR61    0.1214
ATOM   CPZ       CG2R61   0.8129
GROUP   ! NME4
ATOM   NME       NG3P0    -0.8406
ATOM CA1     CG334          -0.35 !
ATOM CA2     CG334          -0.35 !
ATOM CA3     CG334          -0.35 !
ATOM HA11    HGP5            0.25 !
ATOM HA12    HGP5            0.25 !
ATOM HA13    HGP5            0.25 !
ATOM HA21    HGP5            0.25 !
ATOM HA22    HGP5            0.25 !
ATOM HA23    HGP5            0.25 !
ATOM HA31    HGP5            0.25
ATOM HA32    HGP5            0.25
ATOM HA33    HGP5            0.25
FIRST NONE LAST NONE

! TRI-OXY BENZENE
BOND CD2 CG CE1 CD1
BOND CZ CE2
BOND CD1 HD1
BOND CZ OH OH HH
BOND CD2 HD2
BOND CE1 OM1 OM1 CM1 CM1 HM11 CM1 HM12 CM1 HM13
BOND CE2 OM2 OM2 CM2 CM2 HM21 CM2 HM22 CM2 HM23
BOND CG    CB
BOND CB    HB1
DOUBLE CD1 CG CE2 CD2   CZ CE1
! IMIDAZOLE
BOND NIE2   CID2          NID1   CIG           CIE1   NID1
DOUBLE NIE2   CIE1        CID2   CIG
DOUBLE CIG OKET
BOND NID1   CPG             CIE1   CNO
BOND CNO NNO CNO HCN
BOND NNO ON ON HON
BOND CID2 CB
! PARA BENZENE
BOND CPD1 CPG   CPD2 CPG   CPE1 CPD1
BOND CPE2 CPD2 CPZ   CPE1 CPZ   CPE2
BOND CPD1 HPD1 CPD2 HPD2 CPE1 HPE1
BOND CPE2 HPE2 CPZ   NME
! NME4
BOND NME   CA1    NME   CA2    NME   CA3
BOND CA1 HA11   CA1   HA12   CA1   HA13
BOND CA2 HA21   CA2   HA22   CA2   HA23
BOND CA3 HA31   CA3   HA32   CA3   HA33
```

```
BONDS
!V(bond) = Kb(b - b0)**2
!
!Kb: kcal/mole/A**2
!b0: A
!
!atom type Kb b0
!
! HBO (DMHBO+)
! Type1 Type2   Kb          b0
CG2R61  CG2DC3  340.270     1.421     !
CG2DC3  HGA4    321.794     1.117     !
CG25C2  NG2R50  137.916     1.378     !
CG2R53  CG2DC1  293.670     1.448     !
NG2D1   OG311   356.882     1.361     !
CG2R61  NG3P0   276.667     1.481     !

! CYRI (r4CI)
CN7B    NG2R51  220.000     1.456     ! ! CN7B NN2     220.0        1.456

ANGLES
!
!V(angle) = Ktheta(Theta - Theta0)**2
!
!V(Urey-Bradley) = Kub(S - S0)**2
!
!Ktheta: kcal/mole/rad**2
!Theta0: degrees
!Kub: kcal/mole/A**2 (Urey-Bradley)
!S0: A
!
!atom types      Ktheta    Theta0   Kub     S0
!
! HBO (DMHBO+)
CG2R61  CG2DC3  CG25C2  102.947  128.268     !
CG2R61  CG2DC3  HGA4    51.029   115.926     !
CG2R61  CG2R61  CG2DC3  56.566   122.760     !
HGA4    CG2DC3  CG25C2  77.790   111.974     !
CG2R53  NG2R51  CG2R61  60.465   129.532     !
CG2R53  NG2R51  CG2R53  153.217  109.122     !
CG25C2  NG2R50  CG2R53  179.157  108.264     !
NG2R51  CG2R53  CG2DC1  71.071   127.431     !
CG2R53  CG2DC1  HGA4    80.180   115.865     !
CG2R53  CG2DC1  NG2D1   73.720   123.425     !
CG2DC1  NG2D1   OG311   122.588  111.135     !
CG2DC1  CG2R53  NG2R50  115.218  119.191     !
CG2DC3  CG25C2  NG2R50  98.430   129.298     !
CG2R53  CG25C2  NG2R50  264.817  109.292     !
NG2D1   OG311   HGP1    97.338   104.071     !
CG2R61  CG2R61  NG3P0   37.372   118.552     !
CG2R61  NG3P0   CG334   91.377   109.518     !

! CYRI (r4CI)
NG2R51  CN7B    ON6B    110.000  112.000     ! ! ON6B CN7B NN2     110.0       112.0
CG2RC0  NG2R51  CN7B    45.000   126.100     ! ! (CE2)CN5  NN2  C7NB  45.0      126.1
CG2R51  NG2R51  CN7B    45.000   127.600     ! ! (CD1)CN4  NN2  C7NB  45.0      127.6
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```
HN7     CN7B    NG2R51  43.000   111.000      ! ! HN7  CN7B NN2      43.0      111.0
CN7B    CN7B    NG2R51  110.000  111.000      ! ! CN7B CN7B NN2     110.0      111.0
CG1N1   CG2R61  CG2RC0  35.000   120.000      ! ! CG1N1  CG2R61 CG2R61    35.00     120.00

! dummies for linking/terminating
HN5     ON5     HN5     00 0
ON5     HN5     ON5     00 0
P       ON2     P       00 0


DIHEDRALS
!
!V(dihedral) = Kchi(1 + cos(n(chi) - delta))
!
!Kchi: kcal/mole
!n: multiplicity
!delta: degrees
!
!atom types              Kchi    n   delta
!
! HBO (DMHBO+)
NG2R51  CG2R53  NG2R50  CG25C2  0.9660   2  180.00 !
NG2R51  CG2R53  CG25C2  NG2R50  1.0460   2  180.00 !
CG25C2  CG2R53  NG2R51  CG2R53  1.0260   2  180.00 !
HGR61   CG2R61  CG2R61  NG3P0   0.9000   2  180.00 !
NG2R50  CG2R53  CG2DC1  NG2D1   0.9530   1  180.00 !
NG2R50  CG2R53  CG2DC1  NG2D1   0.9430   2  180.00 !
NG2R50  CG2R53  CG2DC1  NG2D1   0.9960   3  180.00 !
CG2R61  CG2R61  CG2DC3  HGA4    0.8790   2  180.00 !
NG2R51  CG2R53  CG2DC1  NG2D1   1.0150   2  180.00 !
NG2R51  CG2R53  CG2DC1  HGA4    0.9550   2  180.00 !
CG2DC3  CG2R61  CG2R61  CG2R61  1.0410   2  180.00 !
CG2R61  NG2R51  CG2R53  CG2DC1  0.9810   2  180.00 !
CG2R61  CG2R61  NG3P0   CG334   1.0100   2  180.00 !
CG2DC1  CG2R53  NG2R50  CG25C2  0.9670   2  180.00 !
CG2R61  CG2DC3  CG25C2  NG2R50  0.9750   2  180.00 !
CG2R61  CG2DC3  CG25C2  CG2R53  1.0380   2  180.00 !
CG2R53  NG2R51  CG2R53  CG2DC1  1.0290   2  180.00 !
OG2D1   CG2R53  NG2R51  CG2R53  0.9910   2  180.00 !
CG2R61  NG2R51  CG2R53  NG2R50  0.9440   2  180.00 !
CG25C2  CG2R53  NG2R51  CG2R61  1.0030   2  180.00 !
CG2DC3  CG25C2  NG2R50  CG2R53  1.0760   2  180.00 !
CG2DC3  CG2R61  CG2R61  HGR61   1.0100   2  180.00 !
CG2R53  CG25C2  NG2R50  CG2R53  1.0280   2  180.00 !
CG2R53  CG2DC1  NG2D1   OG311   0.9870   2  180.00 !
CG2R53  NG2R51  CG2R53  NG2R50  1.0460   2  180.00 !
CG2DC1  NG2D1   OG311   HGP1    1.0130   2  180.00 !
NG2R50  CG2R53  CG2DC1  HGA4    0.9570   2  180.00 !
CG2R61  NG3P0   CG334   HGP5    0.9960   2  180.00 !
HGA4    CG2DC3  CG25C2  NG2R50  0.8620   2  180.00 !
HGA4    CG2DC3  CG25C2  CG2R53  0.9670   2  180.00 !
CG2R61  CG2R61  CG2DC3  CG25C2  0.9470   2  180.00 !
OG301   CG2R61  CG2R61  OG311   0.9710   2  180.00 !
OG2D1   CG2R53  NG2R51  CG2R61  0.9060   2  180.00 !
OG2D1   CG2R53  CG25C2  NG2R50  1.0220   2  180.00 !
HGA4    CG2DC1  NG2D1   OG311   0.9690   2  180.00 !
```

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CG2R61   CG2R61   CG2R61   NG3P0    1.0300    2   180.00 !
CG2R53   NG2R51   CG2R61   CG2R61   1.0000    2   180.00 !

! CYRI (r4CI)
CN7      ON6B     CN7B     NG2R51  0.0       3          0.0 ! CN7   ON6B CN7B NN2        0.0       3          0.0
ON6B     CN7B     NG2R51   CG2RC0  1.1       1        180.0 ! ON6B  CN7B NN2  CN5        1.1       1        180.0 !
ON6B     CN7B     NG2R51   CG2R51  1.1       1          0.0 ! ON6B  CN7B NN2  CN4        1.1       1          0.0 !
CN7B     NG2R51   CG2RC0   CG2RC0  11.0      2        180.0 ! CN7B  NN2  CN5  CN5       11.0      2        180.0 ! adm jr.
CN7B     NG2R51   CG2RC0   CG2R61  11.0      2        180.0 ! CN7B  NN2  CN5  CN5       11.0      2        180.0 ! adm jr.
CN7B     NG2R51   CG2R51   CG2R51  11.0      2        180.0 ! CN7B  NN2  CN4  NN3A      11.0      2        180.0 ! adm jr.
CN7B     NG2R51   CG2R51   HGR52   0.3       2        180.0 ! CN7B  NN2  CN4  HN3        0.3       2        180.0 ! NF
HN7      CN7B     NG2R51   CG2RC0  0.0       3          0.0 ! HN7   CN7B NN2  CN5        0.0       3          0.0 ! NF
CN7      CN7B     NG2R51   CG2R51  0.195     3          0.0 ! HN7   CN7B NN2  CN4        0.195     3          0.0 ! NF
CN7      CN7B     CN7B     NG2R51  0.0       3          0.0 ! CN7   CN7B CN7B NN2        0.0       3          0.0
CN7B     CN7B     NG2R51   CG2RC0  0.3       3          0.0 ! CN7B  CN7B NN2  CN5        0.3       3          0.0 ! NF
CN7B     CN7B     NG2R51   CG2R51  0.0       3        180.0 ! CN7B  CN7B NN2  CN4        0.0       3        180.0 ! NF
HN7      CN7B     CN7B     NG2R51  0.0       3          0.0 ! HN7   CN7B CN7B NN2        0.0       3          0.0
ON5      CN7B     CN7B     NG2R51  0.00      3          0.0 ! NN2   CN7B CN7B ON5        0.000     3          0.0 ! Adenine and cytosine
HN7      CN7B     NG2R51   CG2R51  0.195     3          0.0 ! HN7   CN7B NN2  CN4        0.195     3          0.0 ! NF
CG2R61   CG2R61   CG1N1    NG1T1   0.0000    1   0.00   !
CG2RC0   CG2R61   CG1N1    NG1T1   0.0000    1   0.00   !
CG2R51   CG2RC0   CG2R61   CG1N1   2.9950    2   180.00 !
CG2RC0   CG2RC0   CG2R61   CG1N1   2.9990    2   180.00 !
IMPROPER
!
!V(improper) = Kpsi(psi - psi0)**2
!
!Kpsi: kcal/mole/rad**2
!psi0: degrees
!note that the second column of numbers (0) is ignored
!
!atom types            Kpsi                   psi0
!

NONBONDED nbxmod  5 atom cdiel shift vatom vdistance vswitch -
cutnb 14.0 ctofnb 12.0 ctonnb 10.0 eps 1.0 e14fac 1.0 wmin 1.5
!
!V(Lennard-Jones) = Eps,i,j[(Rmin,i,j/ri,j)**12 - 2(Rmin,i,j/ri,j)**6]
!
!epsilon: kcal/mole, Eps,i,j = sqrt(eps,i * eps,j)
!Rmin/2: A, Rmin,i,j = Rmin/2,i + Rmin/2,j
!
!atom  ignored    epsilon      Rmin/2   ignored   eps,1-4       Rmin/2,1-4
!

END
```

Initial Orientations

The initial orientations for all simulations is shown in the following. For each species except U23, which does not have an internal orientation, two intrahelical ($I_{1,2}$, rotated by 180°) and three extrahelical (E_{1-3}) orientations were constructed. r4CI is highlighted in red, while $DMHBO^+$ is highlighted in grey.

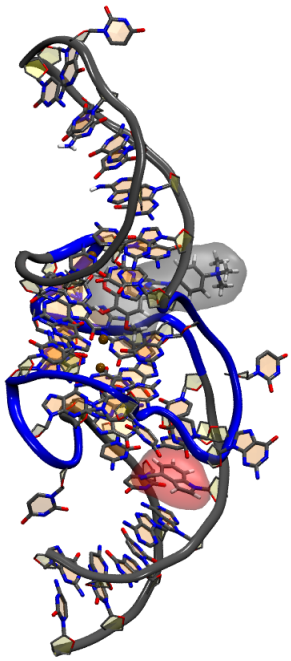

Figure 2: I_1 orientation of the G6 species.

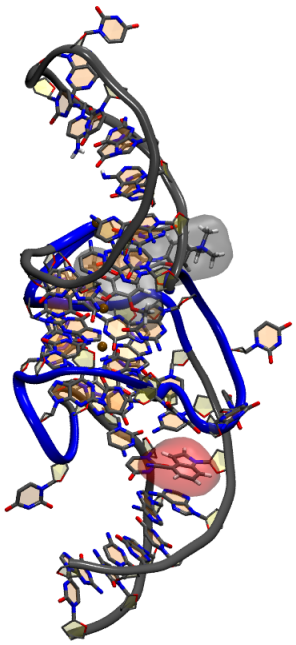

Figure 3: I_2 orientation of the G6 species.

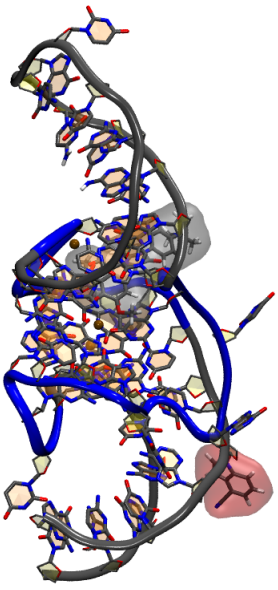

Figure 4: E_1 orientation of the G6 species.

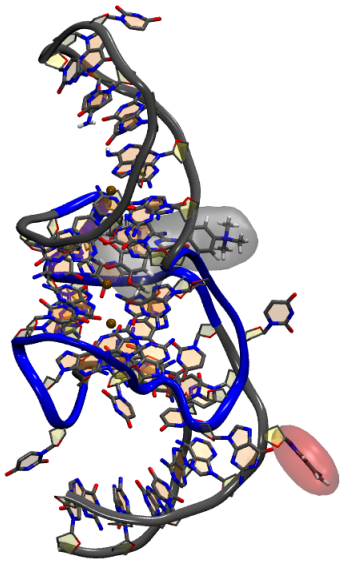

Figure 5: E_2 orientation of the G6 species.

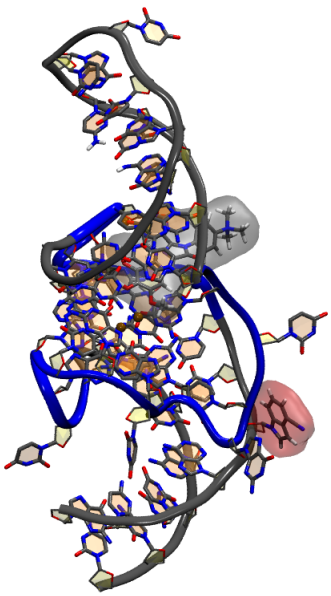

Figure 6: E_3 orientation of the G6 species.

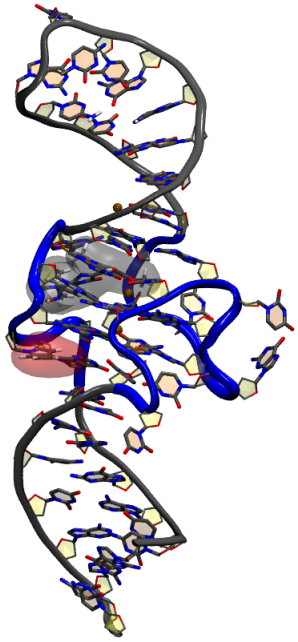

Figure 7: I_1 orientation of the A11 species.

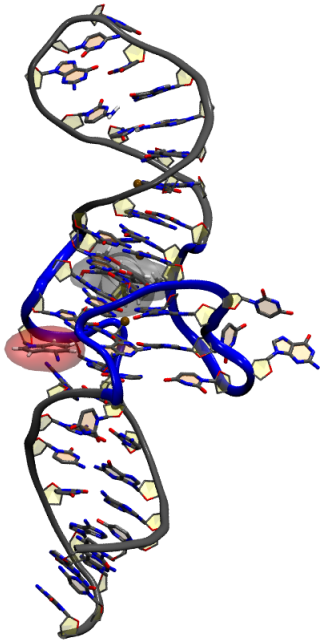

Figure 8: I_2 orientation of the A11 species.

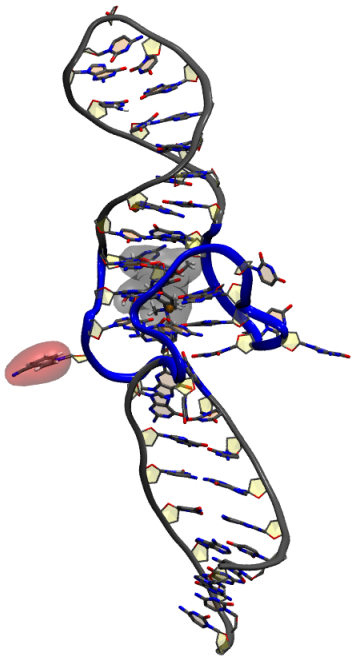

Figure 9: E_1 orientation of the A11 species.

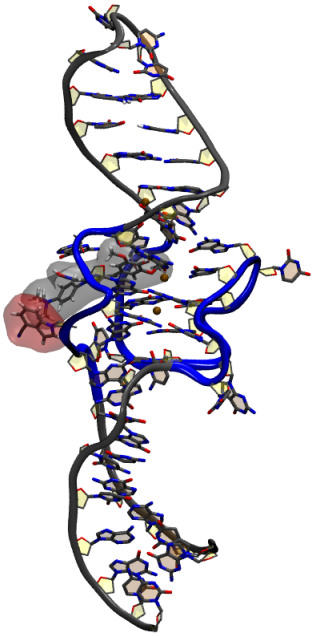

Figure 10: E_2 orientation of the A11 species.

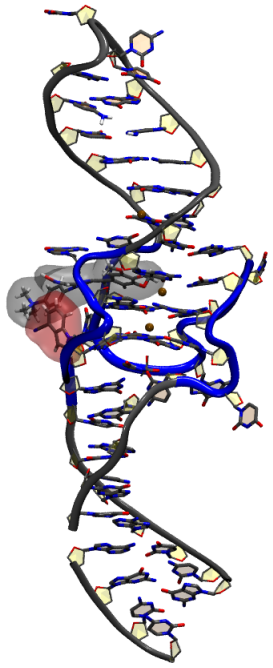

Figure 11: E_3 orientation of the A11 species.

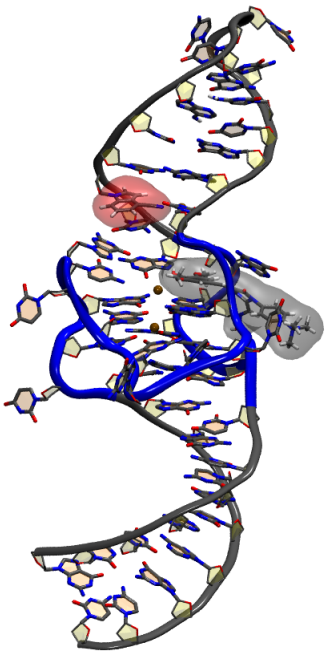

Figure 12: I_1 orientation of the G17 species.

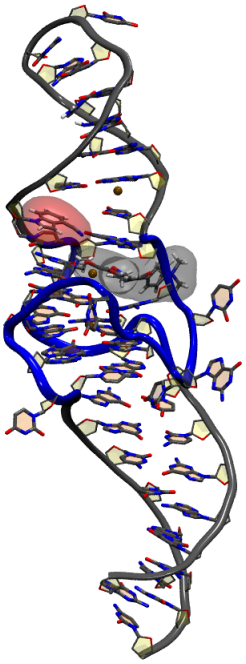

Figure 13: I_2 orientation of the G17 species.

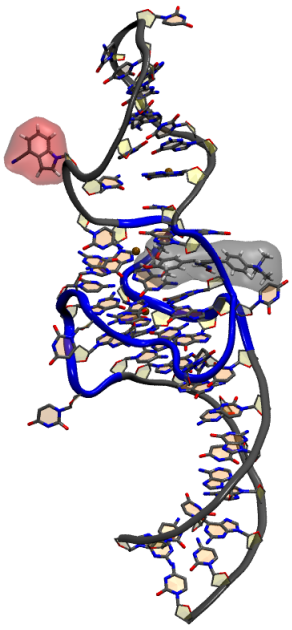

Figure 14: E_1 orientation of the G17 species.

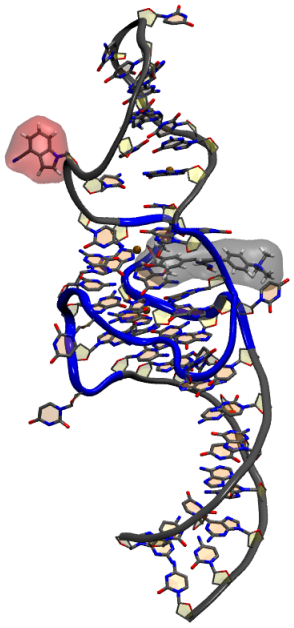

Figure 15: E_2 orientation of the G17 species.

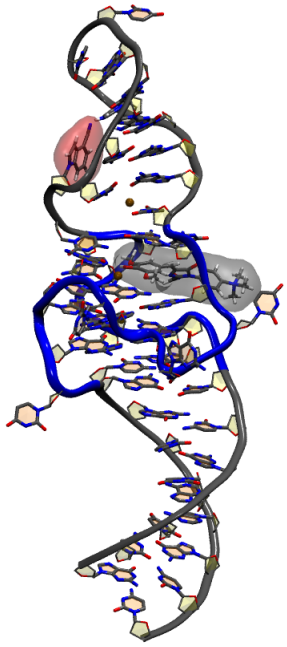

Figure 16: E_3 orientation of the G17 species.

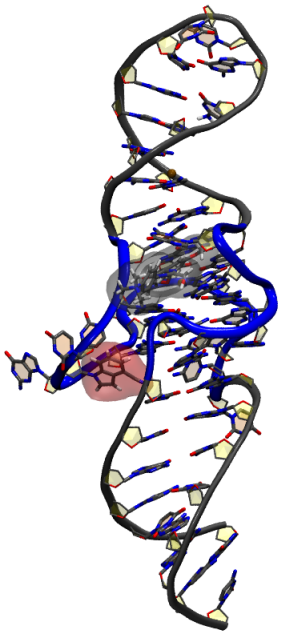

Figure 17: I_1 orientation of the U34 species.

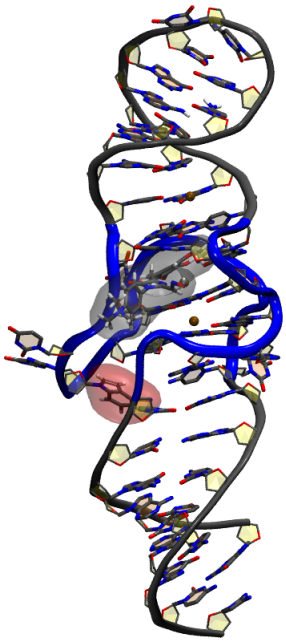

Figure 18: I_2 orientation of the U34 species.

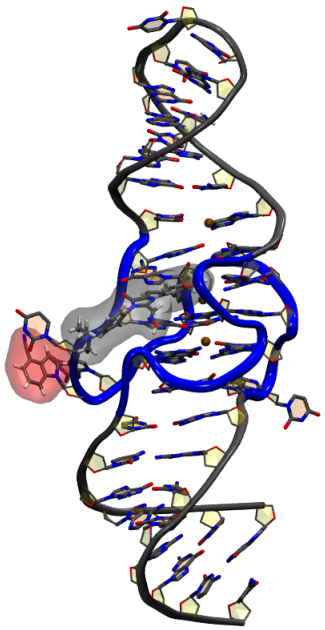

Figure 19: E_1 orientation of the U34 species.

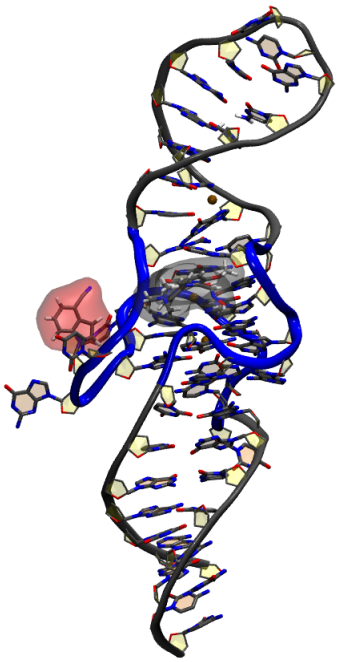

Figure 20: E_2 orientation of the U34 species.

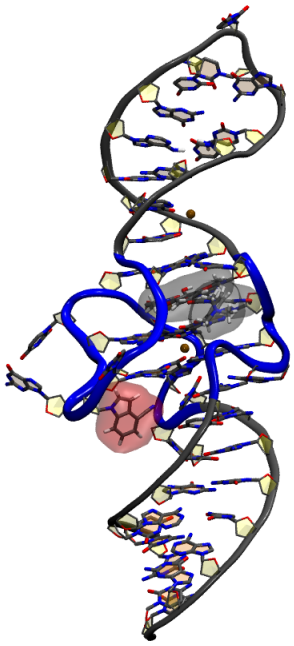

Figure 21: E_3 orientation of the U34 species.

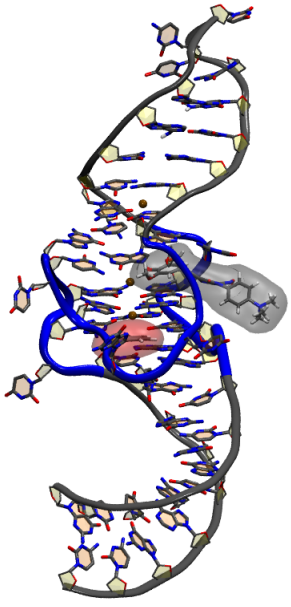

Figure 22: I_1 orientation of the G36 species.

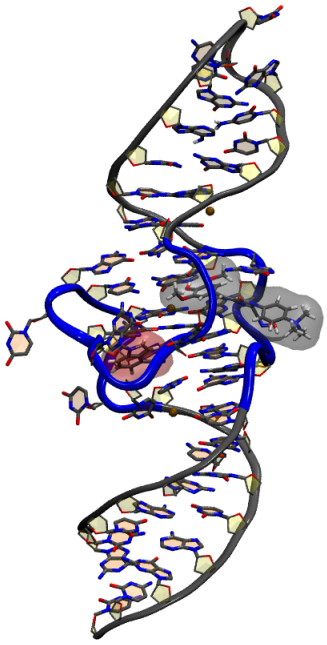

Figure 23: I_2 orientation of the G36 species.

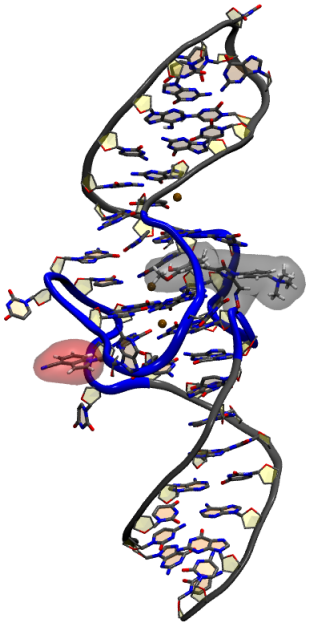

Figure 24: E_1 orientation of the G36 species.

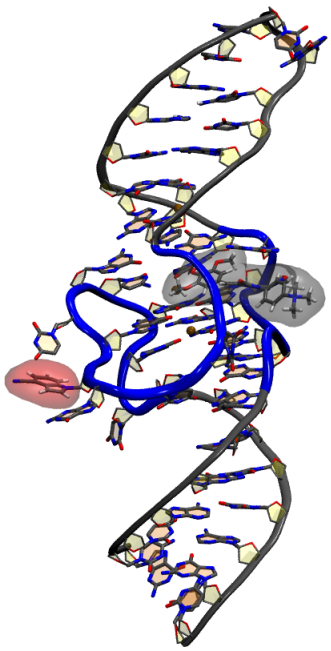

Figure 25: E_2 orientation of the G36 species.

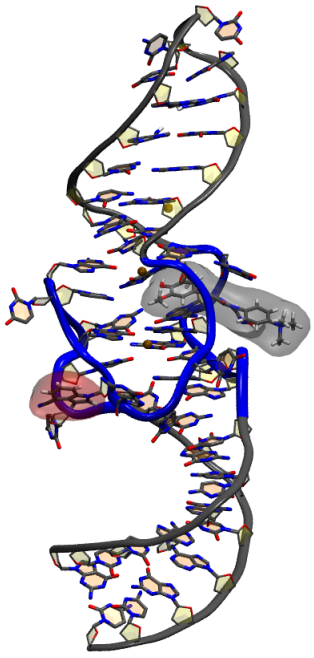

Figure 26: E_3 orientation of the G36 species.

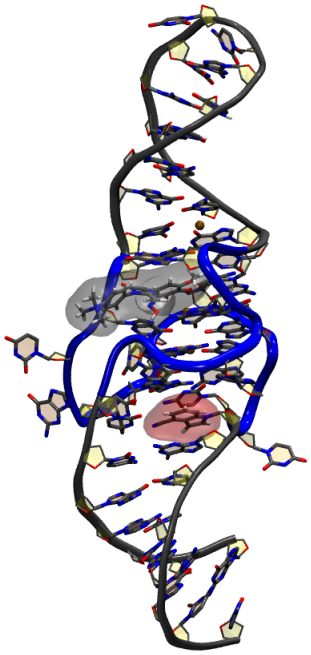

Figure 27: I_1 orientation of the G45 species.

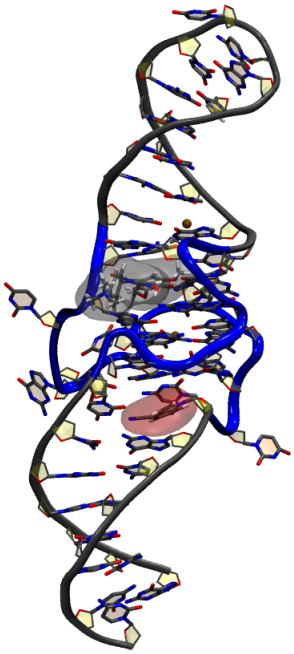

Figure 28: I_2 orientation of the G45 species.

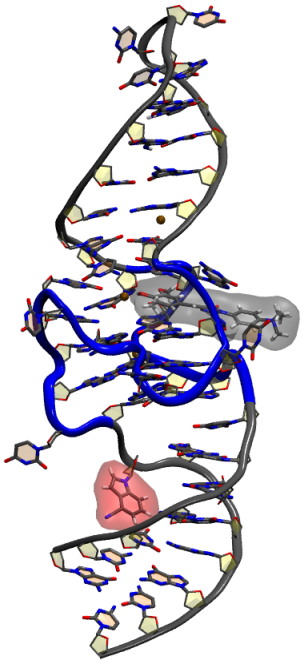

Figure 29: E_1 orientation of the G45 species.

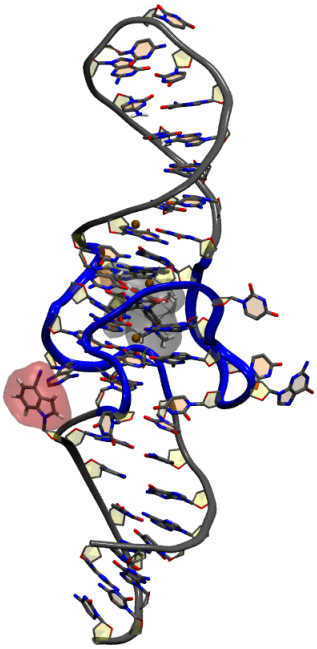

Figure 30: E_2 orientation of the G45 species.

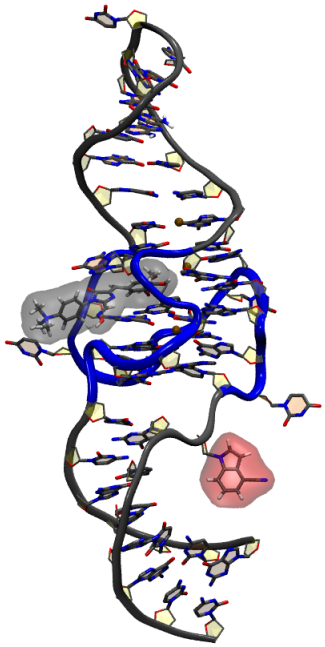

Figure 31: E_3 orientation of the G45 species.

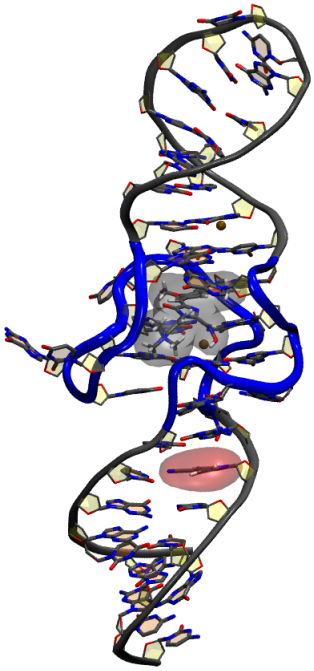

Figure 32: I_1 orientation of the G46 species.

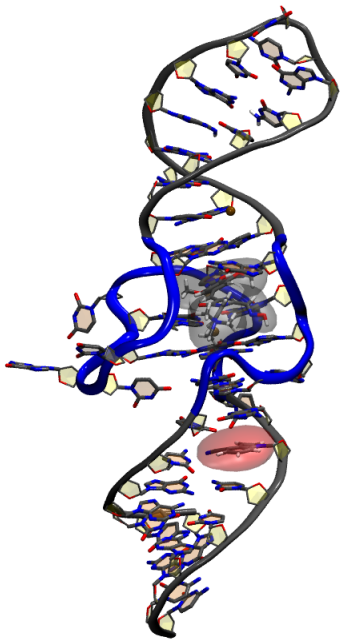

Figure 33: I_2 orientation of the G46 species.

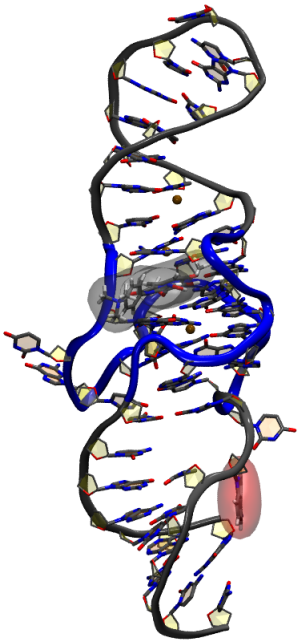

Figure 34: E_1 orientation of the G46 species.

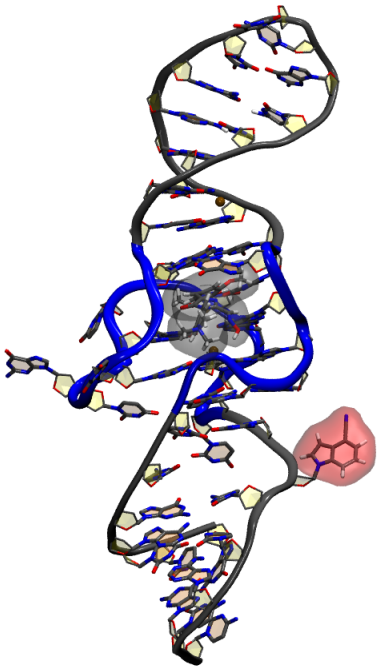

Figure 35: E_2 orientation of the G46 species.

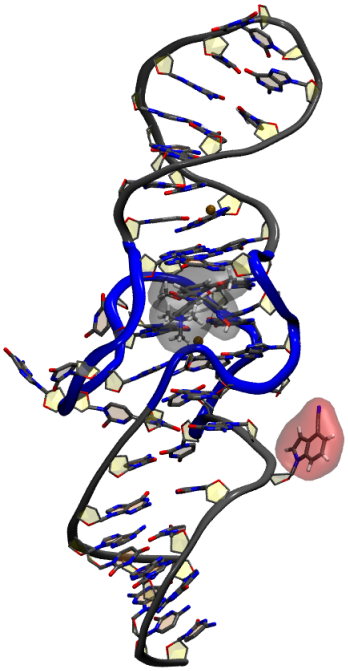

Figure 36: E_3 orientation of the G46 species.

Simulation stability and convergence of trajectories

The RMSD of the heavy atoms as a prime indicator for simulation stability is given for all simulations in the following. The RMSD of the entire aptamer oscilates around 5Å for all simulations. Extrahilical CI exhibits the highest degree of freedom as expected. $DMHBO^+$ oscilates around 3 Å and remains inside the binding site at all times during all simulations.

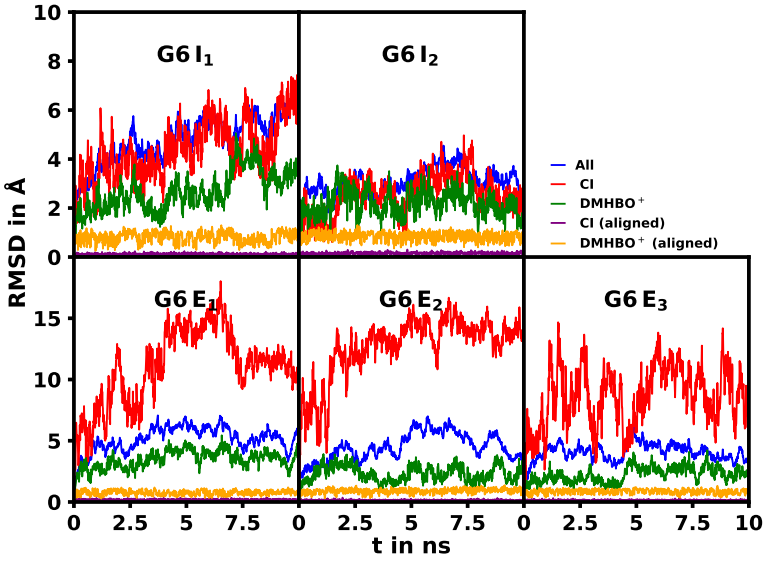

Figure 37: RMSD of the atomic positions in Å of the G6 species simulations.

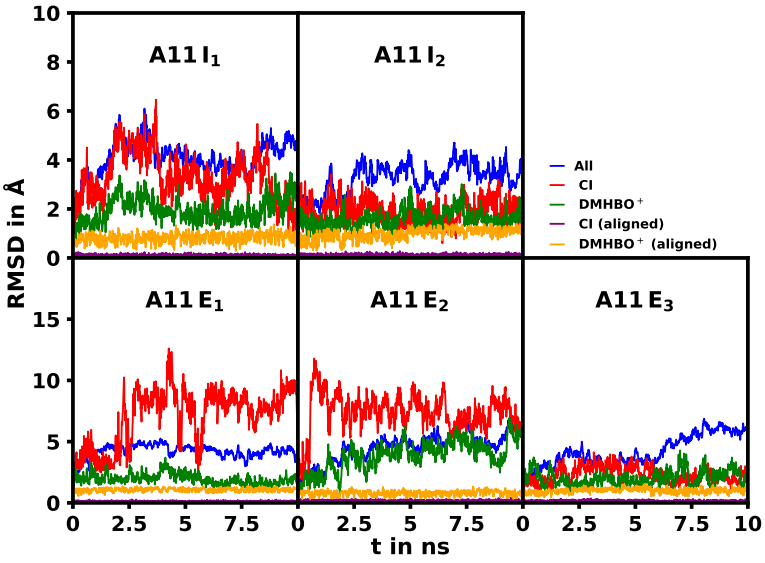

Figure 38: RMSD of the atomic positions in Å of the A11 species simulations.

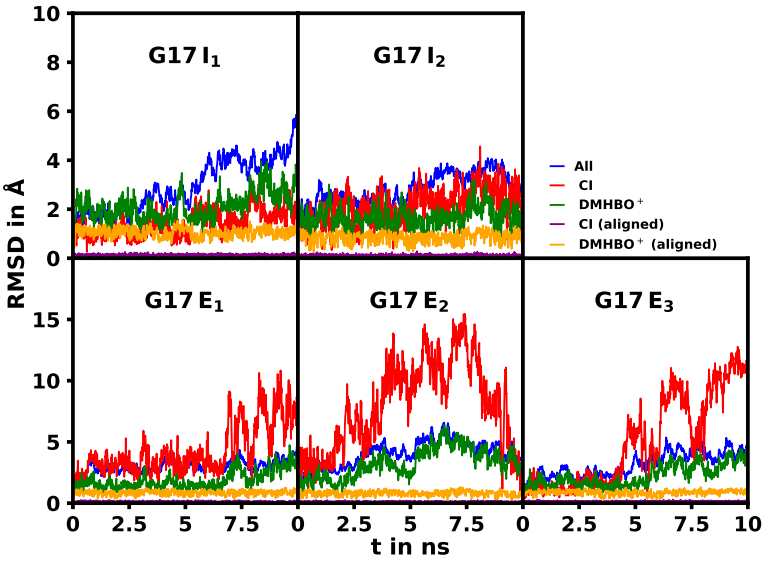

Figure 39: RMSD of the atomic positions in Å of the G17 species simulations.

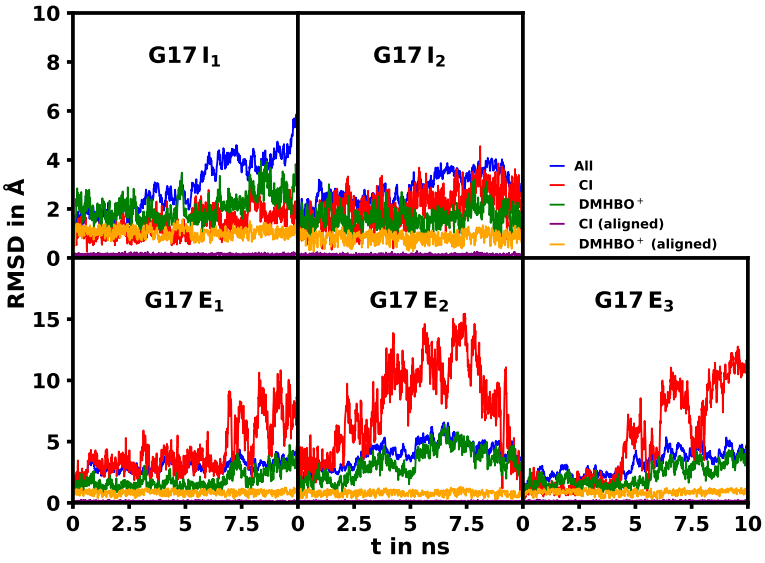

Figure 40: RMSD of the atomic positions in Å of the G17 species simulations.

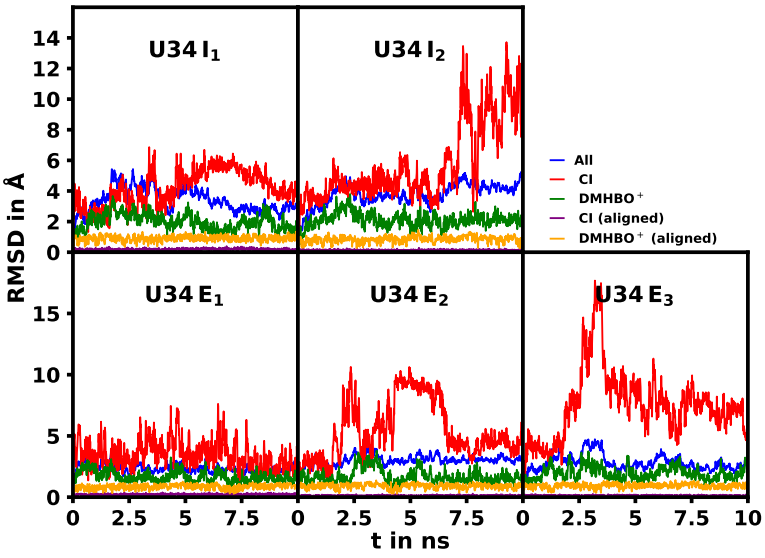

Figure 41: RMSD of the atomic positions in Å of the U34 species simulations.

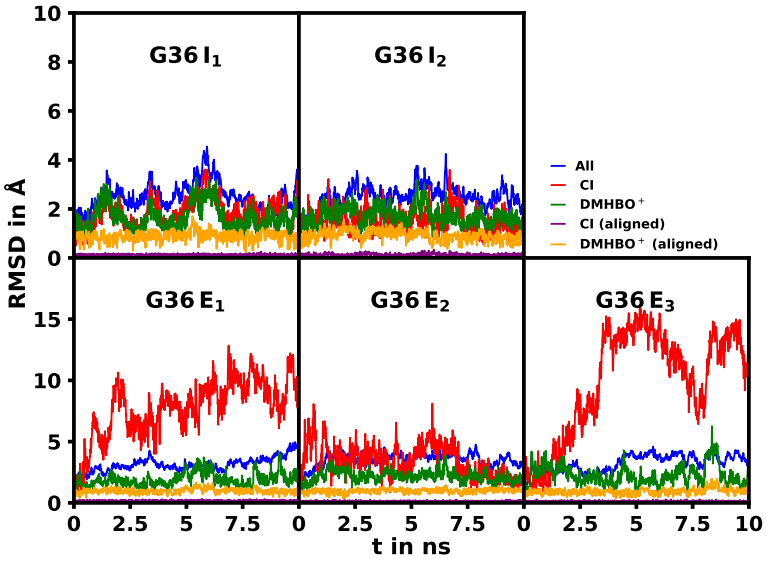

Figure 42: RMSD of the atomic positions in Å of the G36 species simulations.

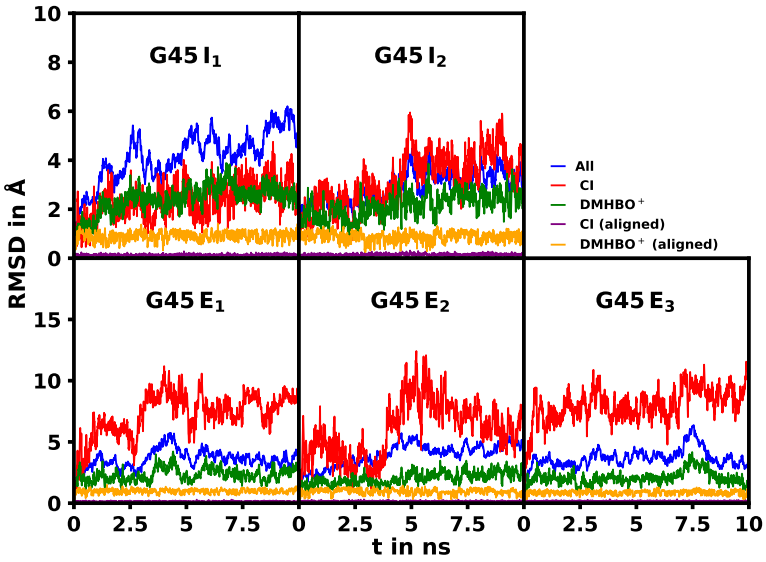

Figure 43: RMSD of the atomic positions in Å of the G45 species simulations.

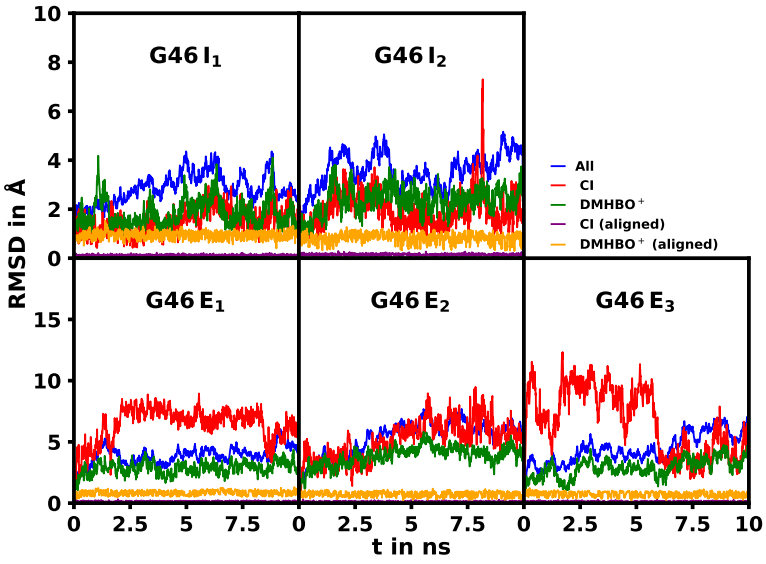

Figure 44: RMSD of the atomic positions in Å of the G46 species simulations.

To assess how well the conformational space of the extrahelical orientations is explored, we have studied whether trajectories cross over into each other. For this, we compare the RMSD in $\mathring{A}$ for all extrahelical trajectories. All trajectories have been aligned along their backbone and for every frame in the E_1 trajectory, we scanned the E_2 and E_3 trajectory for the frame with the lowest RMSD of r4CI compared to E_1 (see Figures 45-51), i.e. the structure most closely resembling the E_1 frame. Examples of crossing points are shown as well. We find that for every species at least 2 trajectories appear to cross over at some point suggesting that the conformational space is reasonably well explored. These trajectories also yield the best results in terms of their R_{DA} values suggesting further that the trjactories we feature are a good representation of the aptamer's structure.

The RMSD of the atomic positions for r4CI shows that E_2 and E_3 of the G6 species are geometrically similar. This is well represented in the two overlayed structures we show.

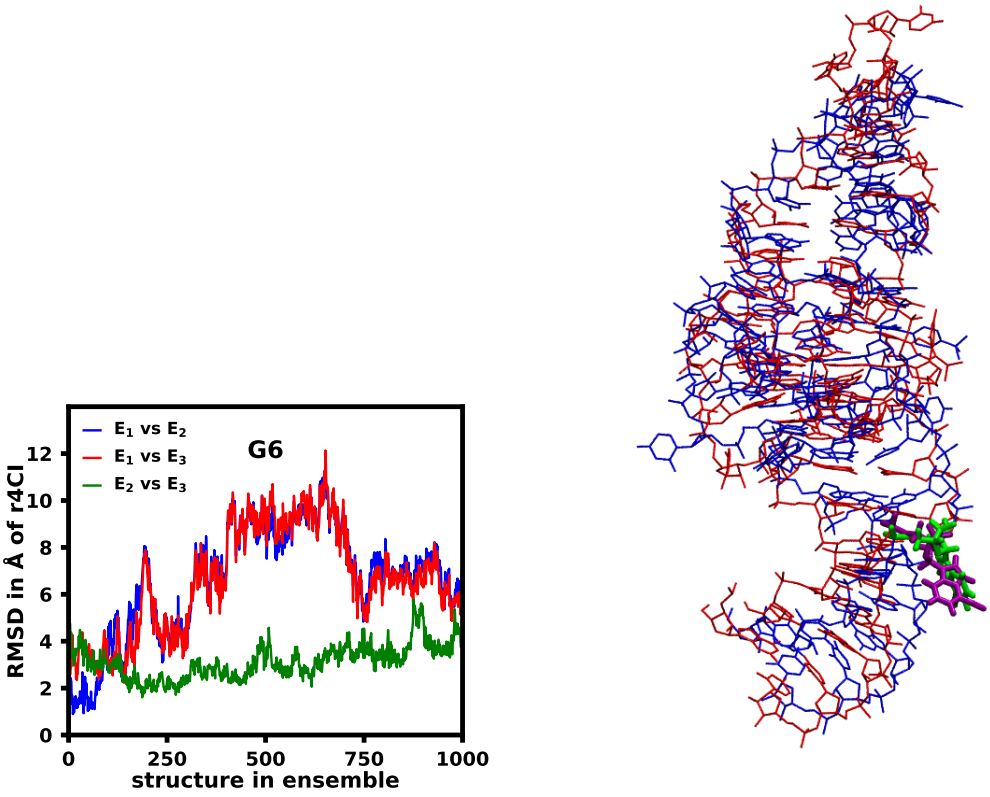

Figure 45: RMSD of the atomic positions in Å for r4CI in the G6 species simulations. The structural representation of structures 209 of E_2 and E_3, shows that aside from the rotation of the cyanoindole group, both structures are geometrically close.

The RMSD of the atomic positions for r4CI shows that E_1 and E_2 of the A11 species are geometrically similar. This is well represented in the two overlayed structures we show.

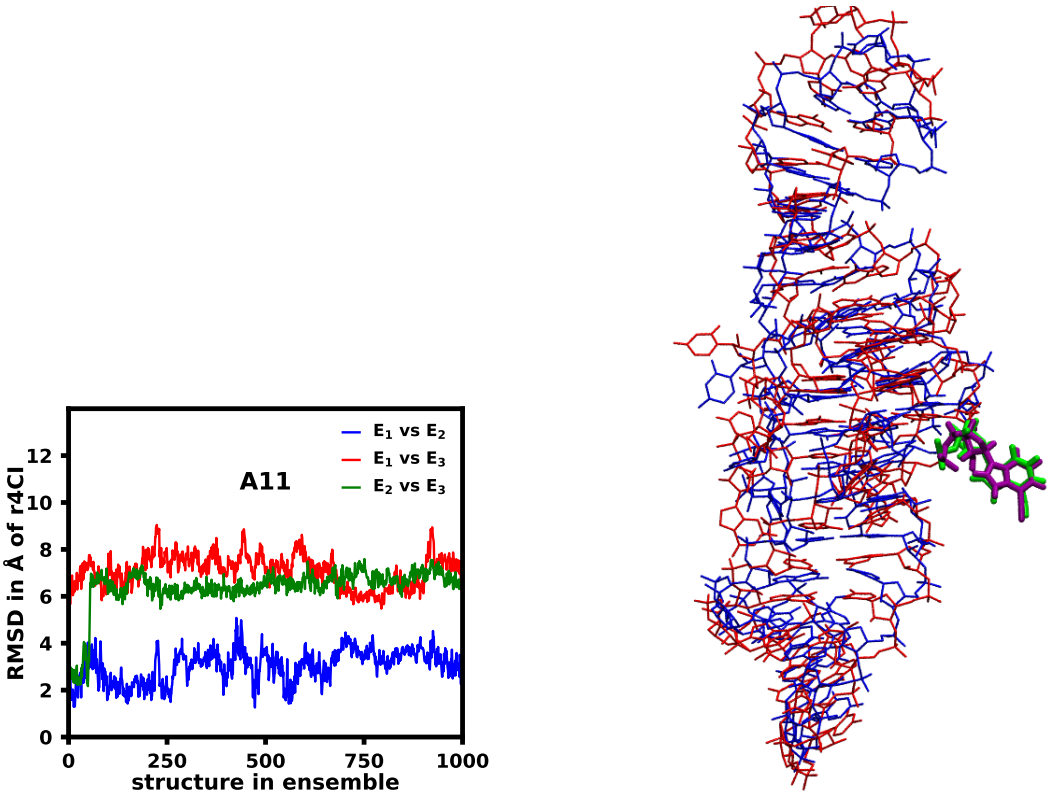

Figure 46: RMSD of the atomic positions in Å for r4CI in the A11 species simulations. The structural representation of structures 547 of E_1 and E_2, shows that both structures are geometrically close.

The RMSD of the atomic positions for r4CI shows that E_1, E_2 and E_3 of the G17 species are geometrically similar. This is well represented in the three overlayed structures we show.

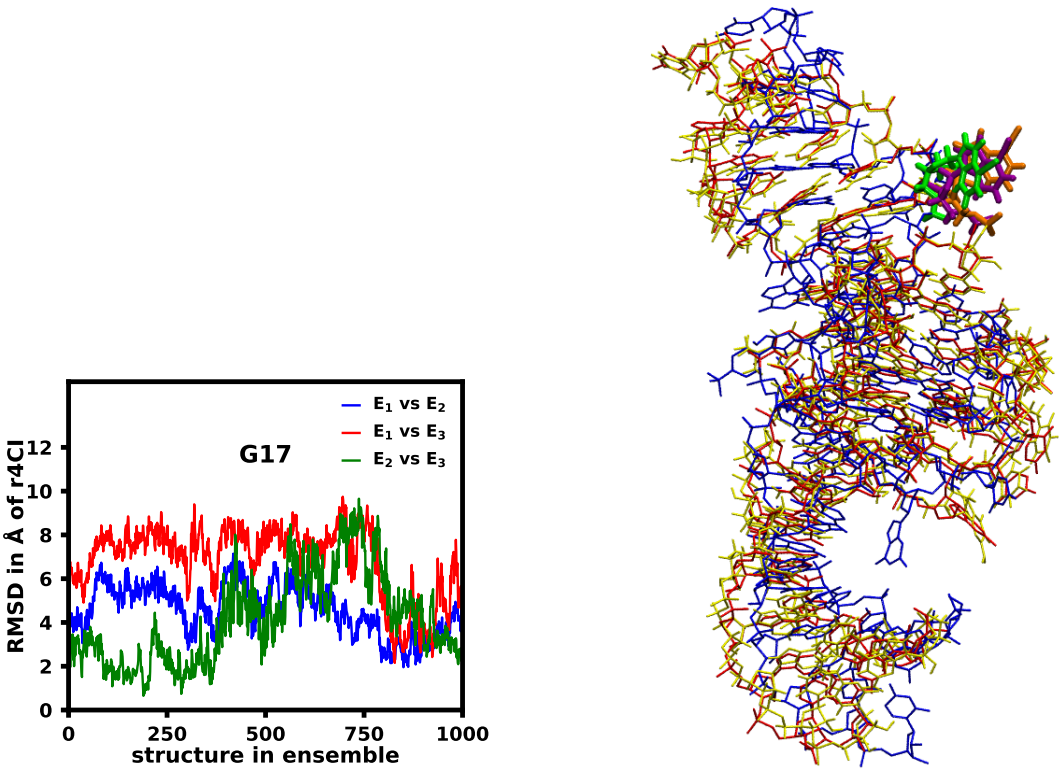

Figure 47: RMSD of the atomic positions in Å for r4CI in the G17 species simulations. The structural representation of structures 846 of E_1, E_2 and E_3, shows that all three structures are geometrically close.

The RMSD of the atomic positions for r4CI shows that E_1, E_2 and E_3 of the U34 species are geometrically similar. This is well represented in the three overlayed structures we show.

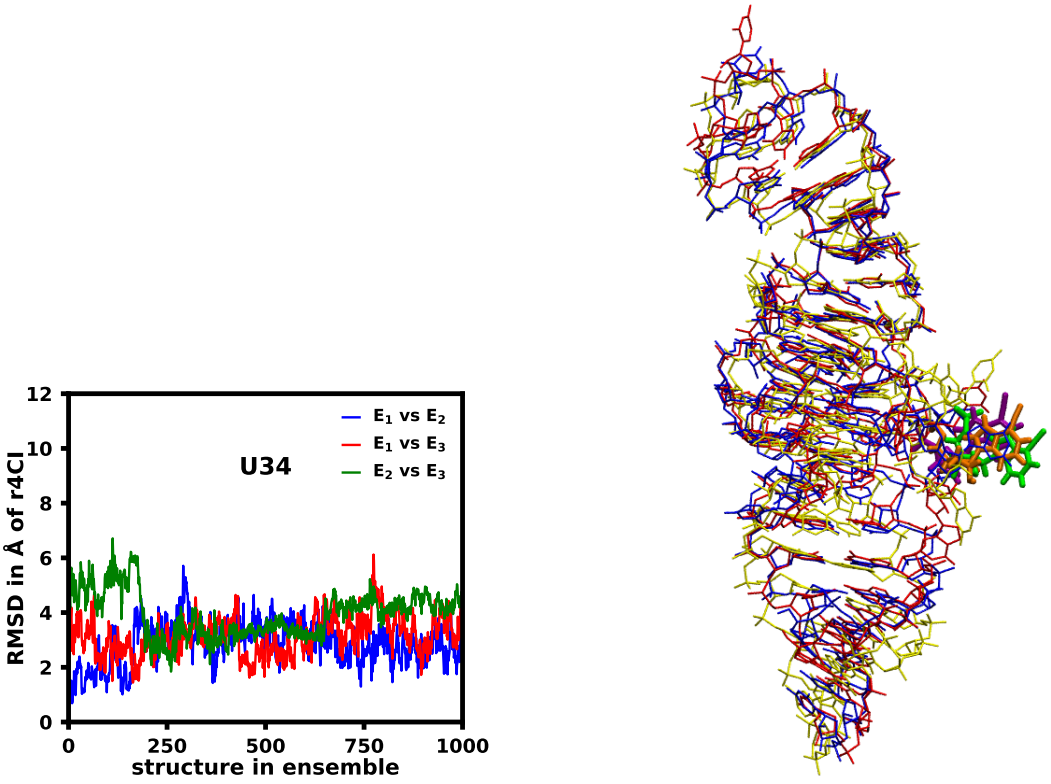

Figure 48: RMSD of the atomic positions in Å for r4CI in the U34 species simulations. The structural representation of structures 231 of E_1, E_2 and E_3, shows that all three structures are geometrically close.

The RMSD of the atomic positions for r4CI shows that E_1, E_2 and E_3 of the G36 species are geometrically similar. This is well represented in the three overlayed structures we show.

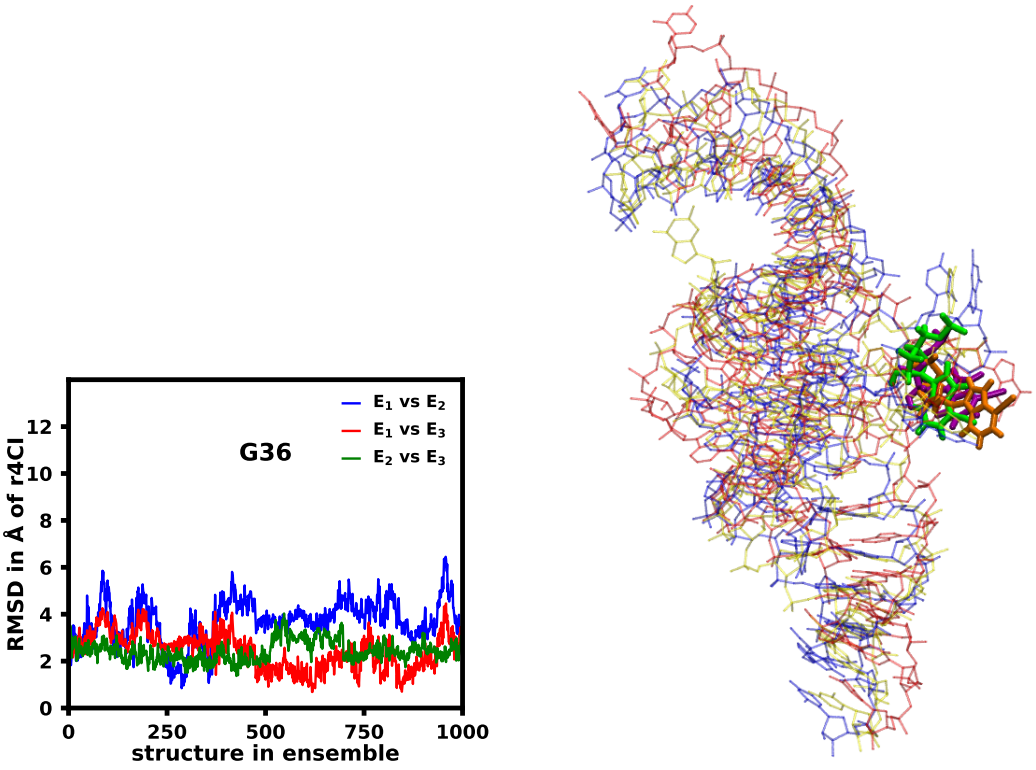

Figure 49: RMSD of the atomic positions in Å for r4CI in the G36 species simulations. The structural representation of structures 120 of E_1, E_2 and E_3, shows that all three structures are geometrically close.

The RMSD of the atomic positions for r4CI shows that E_2 and E_3 of the G45 species are geometrically similar. This is well represented in the two overlayed structures we show.

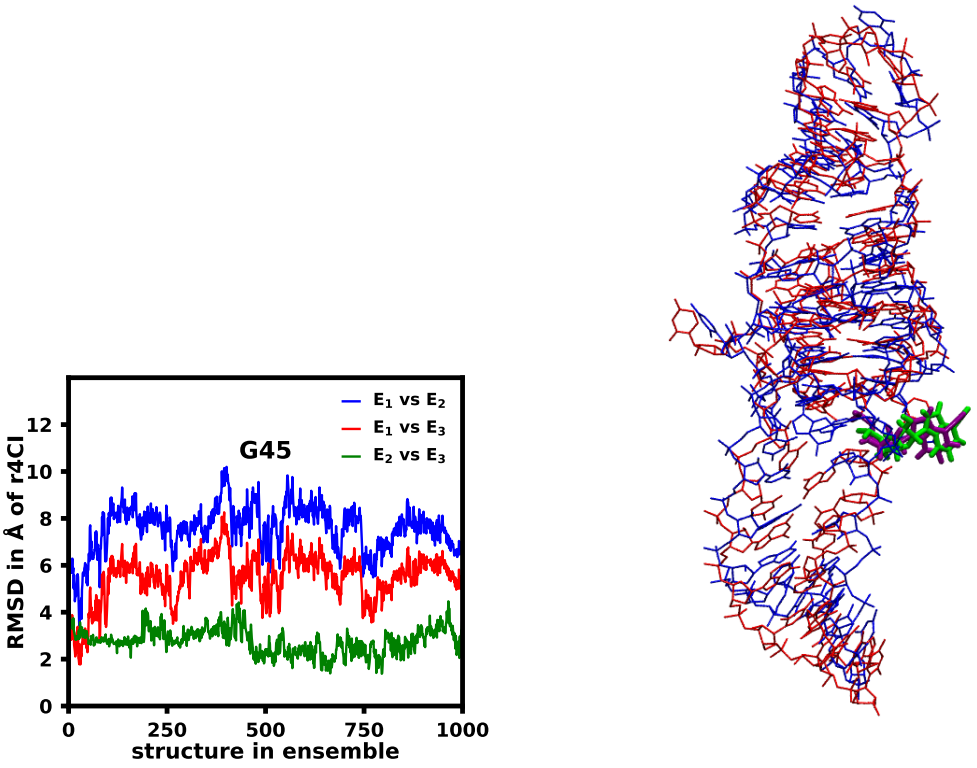

Figure 50: RMSD of the atomic positions in Å for r4CI in the G45 species simulations. The structural representation of structures 761 of E_2 and E_3, shows that both structures are geometrically close.

The RMSD of the atomic positions for r4CI shows that E_1 and E_2 of the G46 species are geometrically similar. This is well represented in the two overlayed structures we show.

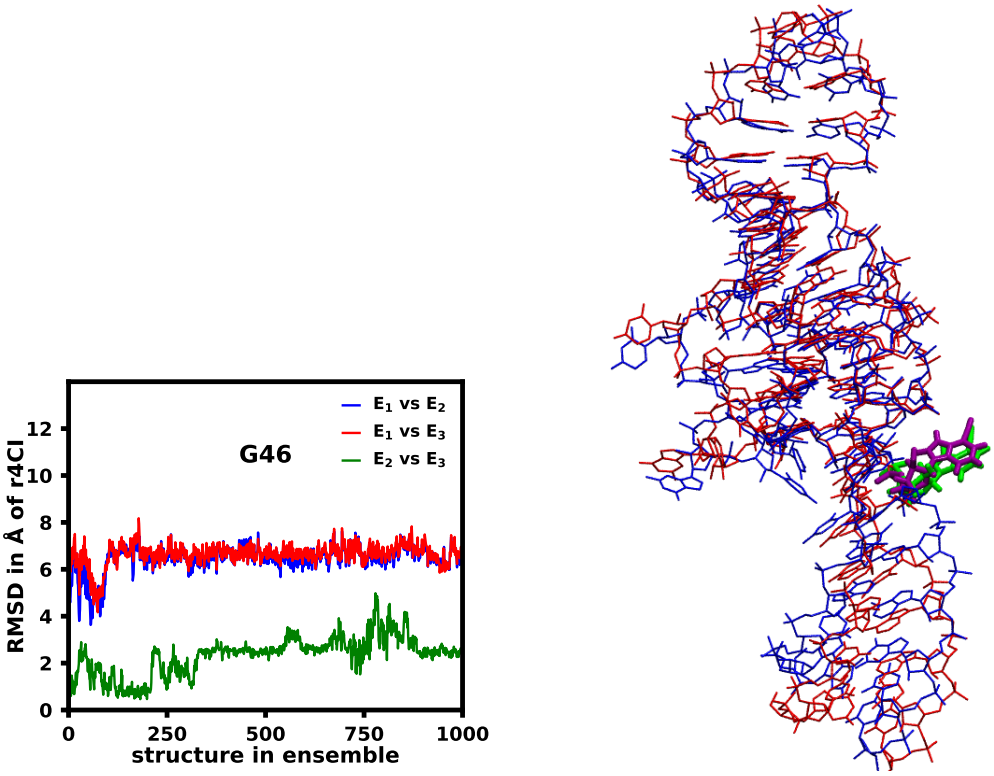

Figure 51: RMSD of the atomic positions in Å for r4CI in the G46 species simulations. The structural representation of structures 234 of E_2 and E_3, shows that both structures are geometrically close.

Chromophoric Distances

The chromophoric distances, R_{DA}, measured over the course of the MD simulations (MD, red) as well as derived from the exciton model (Exc., blue) for all simulations is shown for every simulation run. A summary of the average R_{DA} values for all simulations is shown in table: 1.

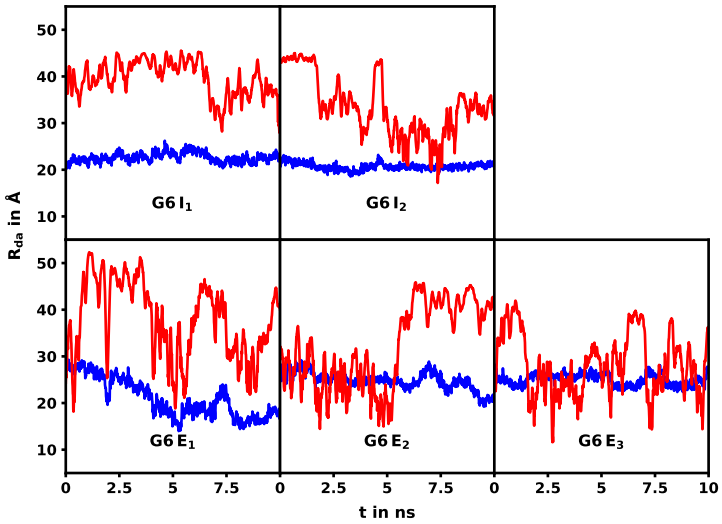

Figure 52: R_{DA} of the G6 species simulations.

Table 1: Spectroscopic data from FRET experiments as well as MD derived theoretical values. The best matches between experiment and simulations are marked with *.

Species	Quantum yield	E_τ	J[a]	R_0 (Å)	κ^2	R_{DA} (Å)	
						MD	Exc.
G6 I_1	0.48	0.36	4.40	36.0	0.53	22.6	39.6
G6 I_2	0.48	0.36	4.40	30.4	0.28	20.7	33.4
G6 E_1	0.48	0.36	4.40	34.2	0.64	21.0	37.6
G6 E_2	0.48	0.36	4.40	29.2	0.31	24.8	32.1
G6 E_3*	0.48	0.36	4.40	29.7	0.13	25.0	28.2
A11 I_1	0.25	0.67	4.90	32.8	0.46	13.0	29.2
A11 I_2	0.25	0.67	4.90	32.3	0.43	13.6	28.7
A11 E_1	0.25	0.67	4.90	19.4	0.04	16.6	19.2
A11 E_2*	0.25	0.67	4.90	22.5	0.04	18.1	20.0
A11 E_3	0.25	0.67	4.90	35.3	0.57	7.5	28.6
G17 I_1	0.07	0.86	4.10	32.6	1.85	16.0	24.1
G17 I_2	0.07	0.86	4.10	33.2	2.09	15.8	24.5
G17 E_1	0.07	0.86	4.10	30.6	1.37	21.2	22.6
G17 E_2	0.07	0.86	4.10	27.5	1.04	23.3	20.2
G17 E_3*	0.07	0.86	4.10	21.0	0.65	20.6	19.4
U23 E_1*	0.49	0.80	4.80	35.8	0.50	31.5	28.1
U34 I_1	0.15	0.70	4.50	28.0	0.44	15.5	24.3
U34 I_2	0.15	0.70	4.50	22.4	0.25	13.8	19.5
U34 E_1	0.15	0.70	4.50	18.9	0.07	19.9	16.4
U34 E_2*	0.15	0.70	4.50	22.0	0.15	21.1	19.5
U34 E_3	0.15	0.70	4.50	26.8	0.40	23.3	16.5
G36 I_1	0.28	0.82	4.40	34.7	0.65	11.7	28.2
G36 I_2	0.28	0.82	4.40	26.2	0.18	13.7	20.3
G36 E_1	0.28	0.82	4.40	38.0	1.14	19.7	29.5
G36 E_2	0.28	0.82	4.40	36.4	0.68	21.0	28.4
G36 E_3*	0.28	0.82	4.40	22.0	0.95	22.0	28.2
G45 I_1	0.10	0.37	4.60	26.1	0.33	14.0	23.2
G45 I_2	0.10	0.37	4.60	24.6	0.26	13.4	26.9
G45 E_1	0.10	0.37	4.60	18.3	0.08	18.5	20.0
G45 E_2	0.10	0.37	4.60	23.6	0.23	22.9	25.9
G45 E_3*	0.10	0.37	4.60	15.4	0.15	22.5	23.2
G46 I_1*	0.28	0.62	4.80	18.9	0.03	17.1	18.3
G46 I_2	0.28	0.62	4.80	18.8	0.03	17.1	18.4
G46 E_1	0.28	0.62	4.80	20.2	0.06	17.4	18.7
G46 E_2	0.28	0.62	4.80	26.3	0.19	17.3	24.2
G46 E_3	0.28	0.62	4.80	28.5	0.36	17.4	26.3

[a] $10^{14} \cdot l \cdot nm^4 \cdot cm^{-1} \cdot mol^{-1}$

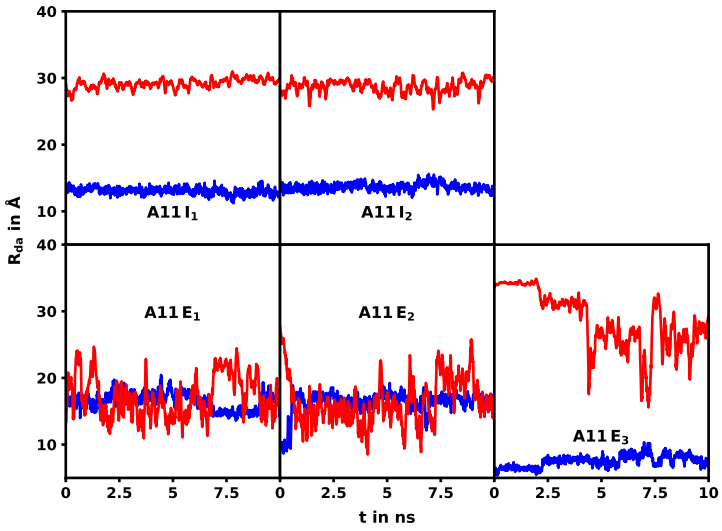

Figure 53: R_{DA} of the A11 species simulations.

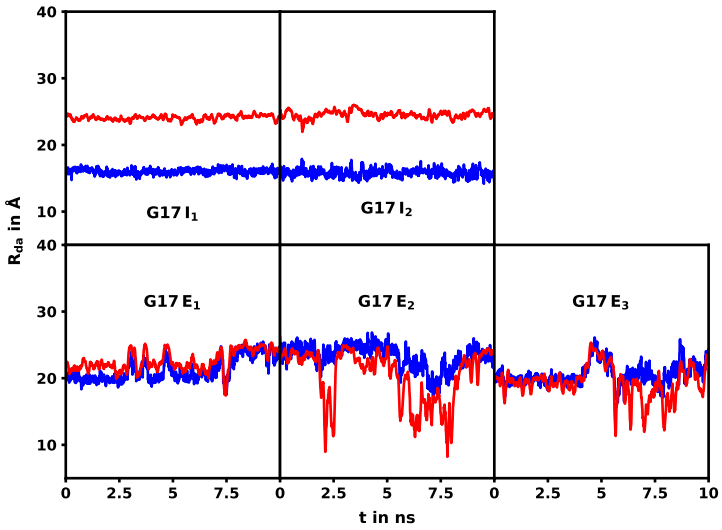

Figure 54: R_{DA} of the G17 species simulations.

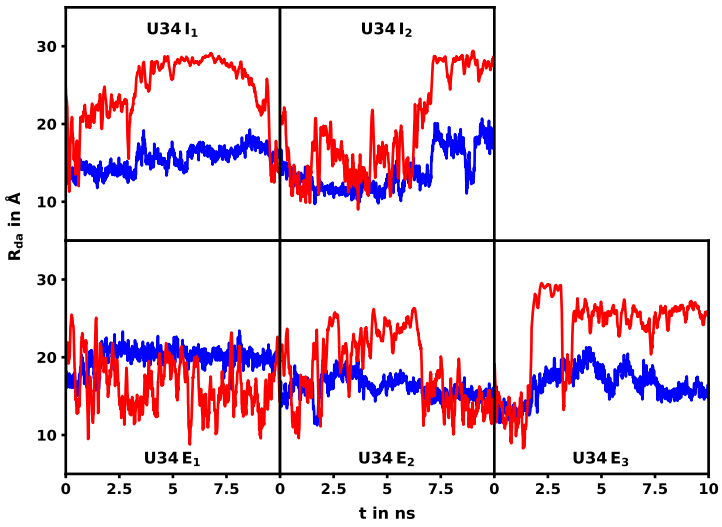

Figure 55: R_{DA} of the U34 species simulations.

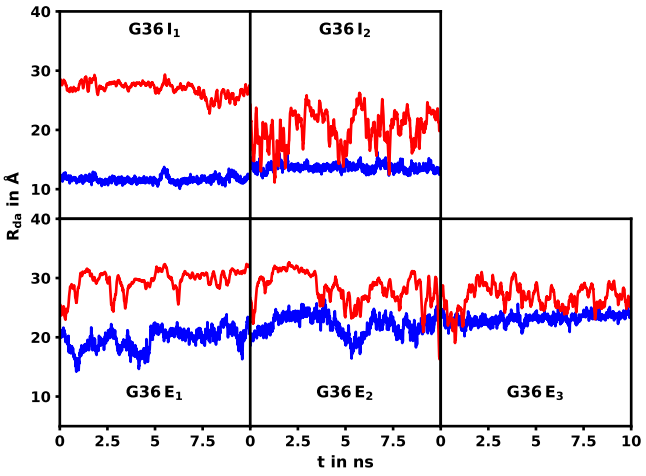

Figure 56: R_{DA} of the G36 species simulations.

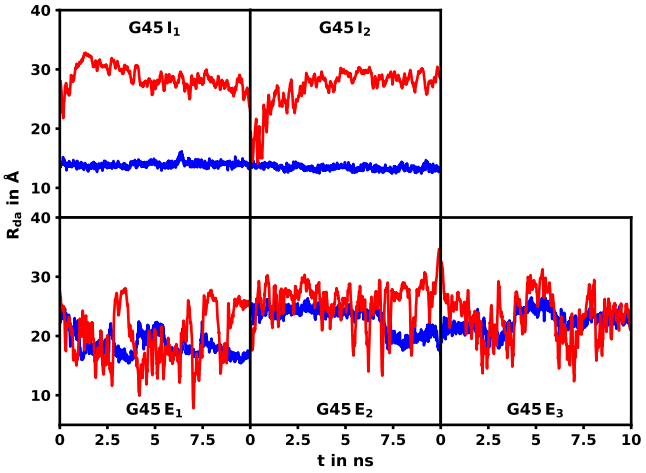

Figure 57: R_{DA} of the G45 species simulations.

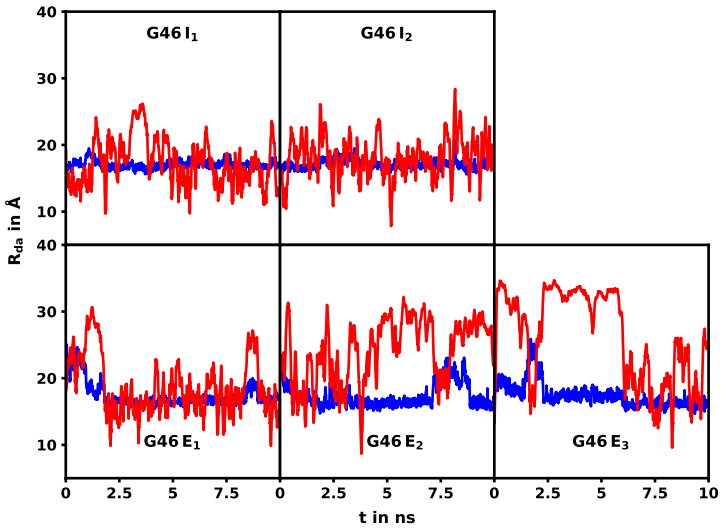

Figure 58: R_{DA} of the G46 species simulations.