Table S1. Detailed RMSD of Induced Fit (IF, RMSD between the bound structure and the most-similar apo-structure to this bound structure) and Conformational Selection (CS, average RMSD between the most-similar apo-structure and other apo-structures).

	Complex	Most-similar Apo	IF RMSD (Å)	CS RMSD (Å)
RNA (no tail)	1	1	1.771	1.367
	2	10	1.498	1.366
	3	9	2.712	1.810
	4	10	1.880	1.366
	5	9	2.123	1.810
	6	1	1.738	1.367
	7	1	1.550	1.367
	8	9	1.776	1.810
	9	1	1.586	1.367
	10	1	1.502	1.367
RNA (with tail)	1	5	3.298	2.954
	2	10	2.570	3.461
	3	9	3.323	3.461
	4	10	2.367	2.719
	5	9	2.471	3.423
	6	1	2.113	2.954
	7	10	2.204	3.204
	8	1	2.612	2.719
	9	10	1.944	3.423
	10	1	1.994	2.954
Protein	1	1	2.139	2.268
	2	9	2.207	2.486
	3	1	1.826	2.268
	4	1	1.798	2.268
	5	7	1.944	2.313
	6	4	1.863	2.380
	7	1	1.723	2.268
	8	7	1.815	2.313
	9	7	2.100	2.313
	10	7	2.397	2.313

Table S2. Detailed populations and standard deviations of all the interactions of population higher than 0.2.

H Bond	Population	*sd*	Hydrophobic	Population	*sd*
18RU_O2P_32THR_N	0.999	0.001	**90VAL_17RG**	0.963	0.080
18RU_O1P_93ALA_N	0.997	0.007	**88ILE_17RG**	0.847	0.164
34GLU_OE2_19RG_N1	0.990	0.023	**90VAL_16RC**	0.401	0.329
19RG_O6_34GLU_N	0.982	0.017	**91PRO_16RC**	0.275	0.378
18RU_O2P_32THR_OG1	0.981	0.024			
34GLU_OE2_6RG_O2'	0.967	0.086			
19RG_O6_33ASN_N	0.929	0.046			
17RG_N7_30LYS_NZ	0.858	0.284			
30LYS_NZ_6RG_O2'	0.856	0.242			
6RG_O2'_30LYS_NZ	0.856	0.242	**Electrostatic**	**Population**	**sd**
34GLU_OE2_19RG_N2	0.833	0.083	**29LYS_6RG**	1.000	0.000
7RA_O2P_30LYS_NZ	0.830	0.249	**30LYS_7RA**	1.000	0.000
5RU_O2P_41ARG_NH2	0.818	0.274	**37LYS_4RC**	1.000	0.000
4RC_O1P_41ARG_NH2	0.773	0.153	**37LYS_5RU**	1.000	0.000
6RG_O6_33ASN_ND2	0.719	0.121	**79LYS_18RU**	1.000	0.000
18RU_O4_54ASP_N	0.715	0.180	**30LYS_18RU**	1.000	0.000
17RG_O6_30LYS_NZ	0.713	0.305	**29LYS_5RU**	0.999	0.002
5RU_O2P_41ARG_NH1	0.672	0.224	**41ARG_5RU**	0.998	0.005
5RU_O1P_41ARG_NH1	0.670	0.199	**41ARG_4RC**	0.998	0.005
6RG_O3'_30LYS_NZ	0.669	0.258	**37LYS_3RU**	0.993	0.010
19RG_O6_32THR_N	0.574	0.152	**79LYS_17RG**	0.959	0.036
34GLU_OE1_6RG_O2'	0.541	0.169	**29LYS_7RA**	0.957	0.121
18RU_O4_79LYS_NZ	0.533	0.160	**30LYS_6RG**	0.938	0.134
6RG_O6_37LYS_NZ	0.511	0.331	**41ARG_6RG**	0.925	0.188
4RC_O2P_37LYS_NZ	0.468	0.254	**27LYS_6RG**	0.855	0.114
6RG_O2P_29LYS_NZ	0.398	0.190	**37LYS_6RG**	0.841	0.232
4RC_O5'_41ARG_NH2	0.378	0.154	**41ARG_3RU**	0.621	0.126
54ASP_O_18RU_N3	0.273	0.367	**30LYS_8RC**	0.575	0.194
4RC_O3'_41ARG_NH2	0.202	0.104	**27LYS_7RA**	0.371	0.203

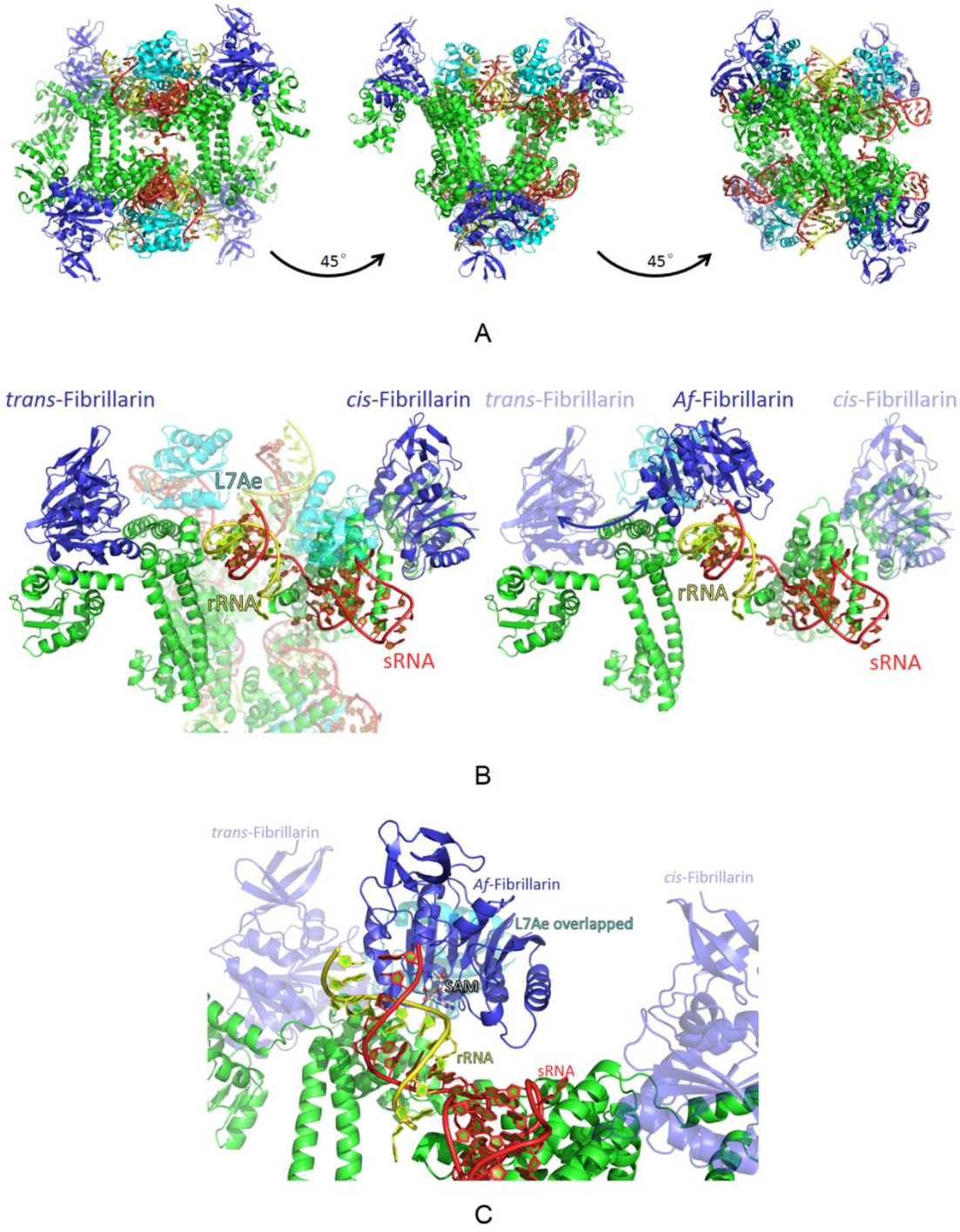

Figure S1. Structures of diRNP and cross-RNP models. (A) The diRNP model was manually constructed based on previous EM results and Xue's docking. (B and C) In diRNP model, fibrillarin (blue) has "*cis*" position and "*trans*" position, which are both far away from substrate rRNA (yellow). The transition of fibrillarin from "*trans*" position to "*Af*" position enables the methylation catalyse (Methyl donor, SAM in ball-stick representation, is very close to substrate rRNA). However, L7Ae (cyan) is overlapped by fibrillarin, indicating it also has a reposition. The reposition of fibrillarin might be under the hinge motion of sRNA, as well as L7Ae. Additionally, K-turn sRNA is in red cartoon representation.

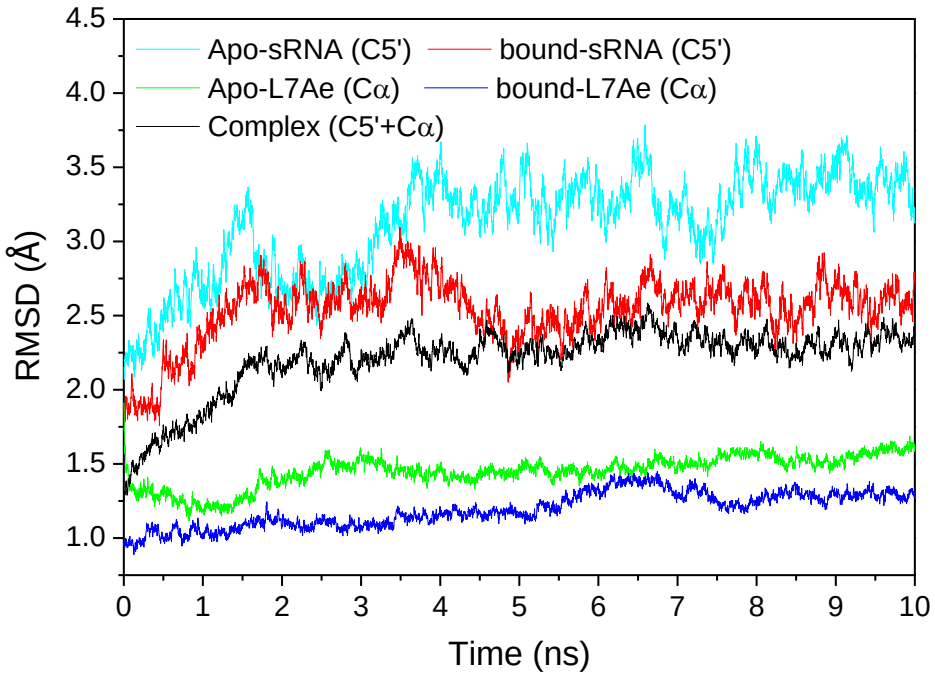

Figure S1. RMSD to the start structure at 298K for bound (black) and apo-sRNA (red) and apo-L7Ae (blue).

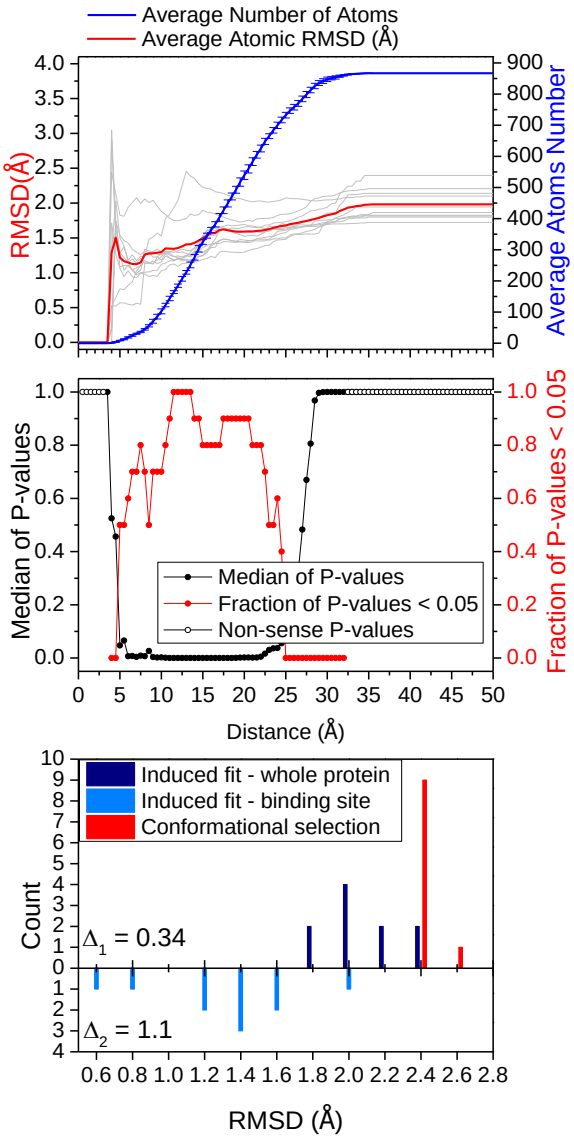

Figure S2. Binding mechanism evaluation for L7Ae binding.

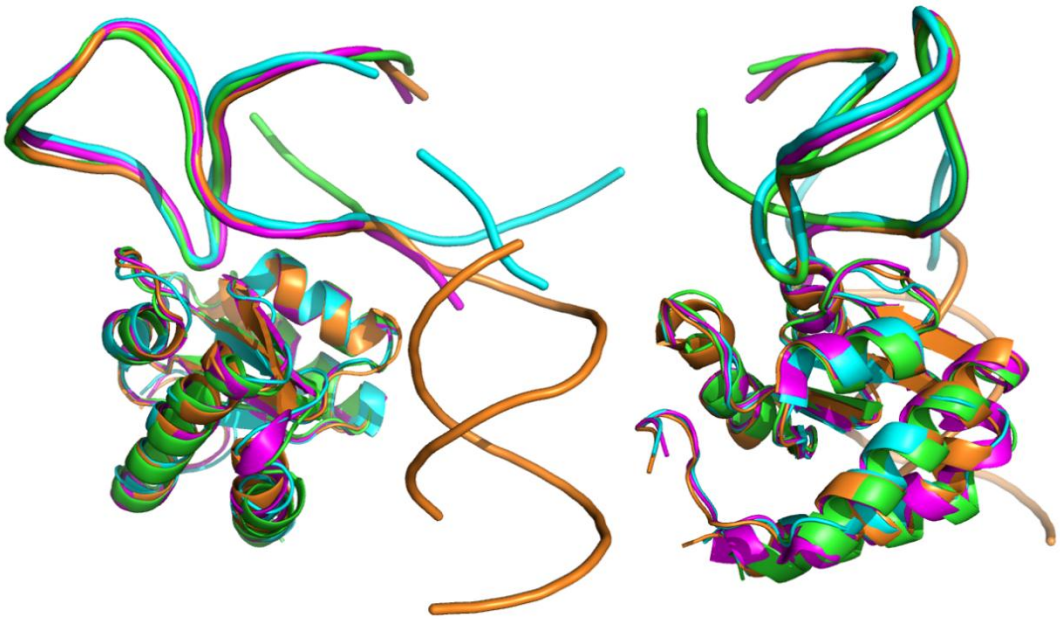

Figure S3. Alignment of 1RLG (green), 3NMU (orange), 3NVK (cyan) and 3NVI (magenta), basing on all Cα atoms, shown in front-sight and lateral-sight.

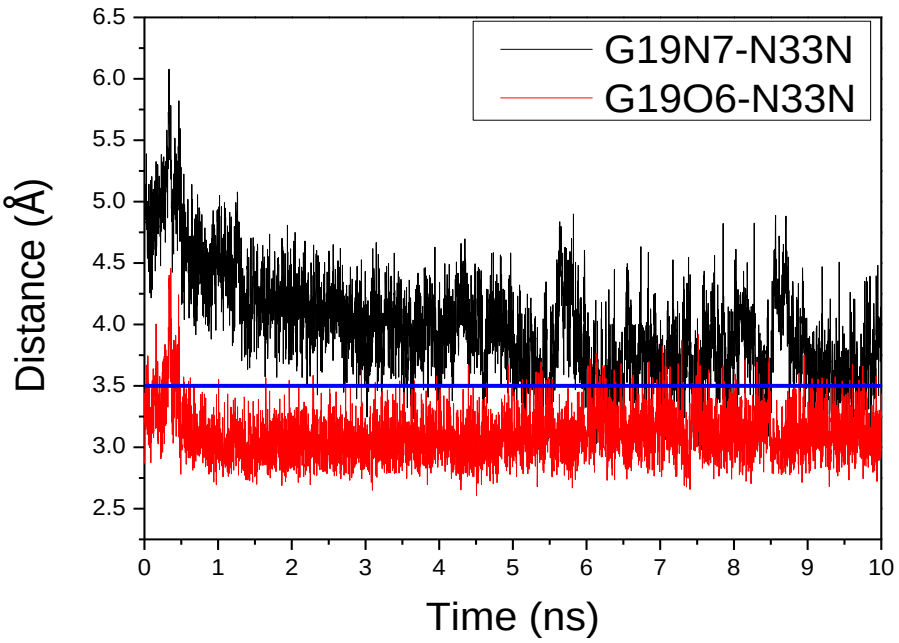

Figure S4. Distances *vs.* time of two hydrogen bonds both between G19 and N33, red line for G19O6-N33N, black for G19N7-N33N, and blue line for the hydrogen bond threshold.

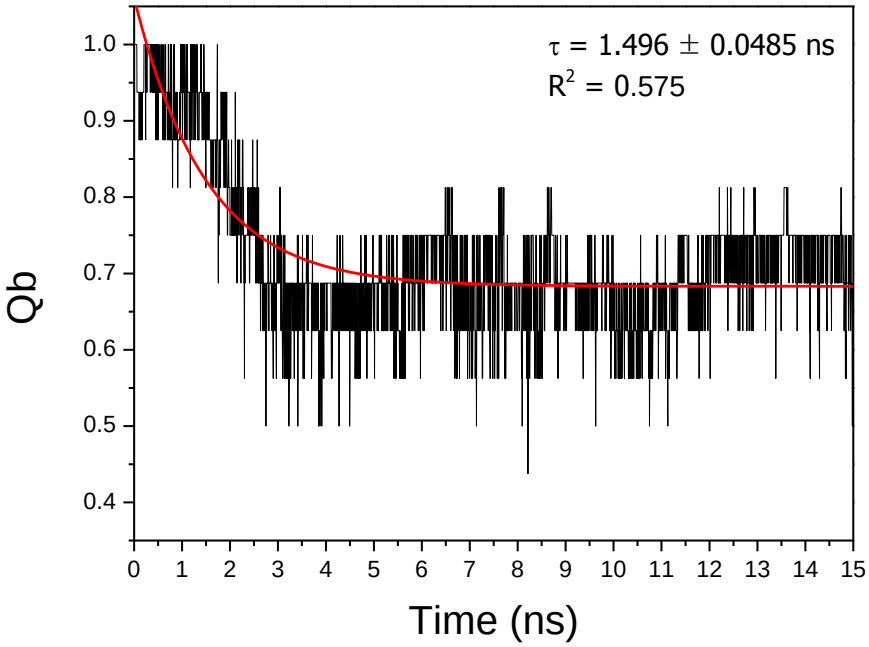

Figure S5. Time evolution of Qb for U18G mutant complex. Fitting of first order exponential function was shown in red line. Unfolding half time and correlation coefficient were also listed.

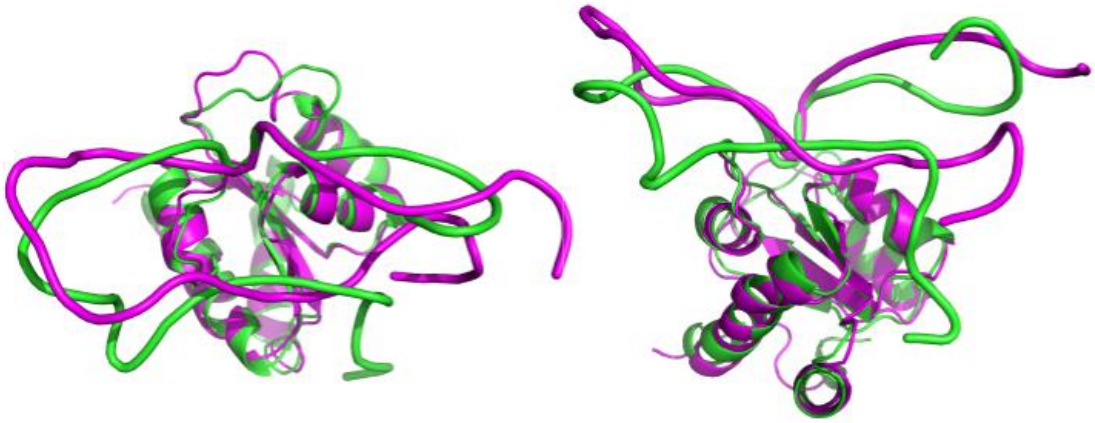

Figure S6. Head view and lateral view of alignment of average structure in transition state (TS) ensembles. Green for U18G mutant bound sRNA and magenta for WT complex.