Table S1. Chemical shifts (in p.p.m.) of the non-exchangeable proton and protonated carbon resonances of ψ_{32}-ACSL RNA hairpin.

Residue	H6/ H8	H5/ H2	H1'	H2'	H3'	H4'	H5'/ H5''	C6/ C8	C5/ C2	C1'	C2'	C3'	C4'	C5'
G_{27}	7.99	-	5.74	4.84	4.54	4.34	4.01, 3.89	[a]n.a.	-	n.a.	n.a.	n.a.	n.a.	n.a.
G_{28}	7.47	-	5.90	4.64	4.56	4.51	4.49, 4.19	136.74	-	92.86	75.65	72.81	82.27	65.77
G_{29}	7.17	-	5.79	4.62	4.48	4.45	4.48, 4.06	135.98	-	92.87	75.69	72.82	81.99	65.41
G_{30}	7.10	-	5.75	4.62	4.42	4.47	4.41, 4.03	136.17		93.12	75.53	73.14	82.23	66.05
A_{31}	7.68	7.74	5.99	4.65	4.58	4.47	4.53, 4.08	139.63	154.26	93.15	75.69	72.56	82.09	64.86
ψ_{32}	6.92	-	4.56	4.10	4.28	4.14	4.37, 3.94	140.05		82.78	75.16	73.65	79.91	65.33
U_{33}	7.51	5.25	5.70	4.31	4.36	4.31	4.40, 4.02	141.49	103.83	93.29	76.23	73.25	82.86	65.02
G_{34}	7.95	-	5.49	4.70	4.56	4.39	4.15, 4.01	139.43		88.67	74.97	77.54	84.82	66.93
A_{35}	8.09	8.01	5.49	4.35	4.56	3.69	3.77, 3.51	141.95	155.17	88.67	77.47	77.54	84.82	67.80
A_{36}	7.88	7.91	5.72	4.74	4.81	4.44	4.10, 3.97	141.16	155.06	89.13	75.57	77.00	84.78	68.59
A_{37}	8.18	7.19	5.76	4.80	4.65	4.59	4.35, 4.30	141.51	153.46	89.36	75.25	75.76	84.34	68.84
A_{38}	7.77	7.86	5.75	4.41	4.36	4.56	4.33, n.a.	139.90	154.09	93.00	75.29	73.82	83.06	68.07
U_{39}	7.61	4.92	5.46	4.29	4.41	4.39	4.51, 4.05	141.64	102.60	93.49	75.33	72.10	82.11	64.26
C_{40}	7.88	5.54	5.58	4.27	4.45	4.40	4.49, 4.04	141.80	97.33	93.83	75.58	72.22	81.90	64.62
C_{41}	7.79	5.43	5.43	4.34	4.45	4.37	4.52, 4.03	141.63	97.44	94.09	75.37	72.06	81.99	64.42
C_{42}	7.80	5.45	5.46	4.24	4.47	4.35	4.53, 4.02	141.60	97.44	94.35	75.53	71.91	82.07	64.30
C_{43}	7.66	7.83	5.71	3.98	4.15	4.14	4.46, 3.99	141.88	97.97	92.87	77.48	69.63	83.66	65.13

The ^{1}H chemical shifts were measured at 25 °C and pH 6.8 and are referenced relative to DSS (0.00 ppm) and the ^{13}C chemical shifts set using the spectrometer frequency and the ^{1}H and ^{13}C gyromagnetic ratios. The uncertainties in the chemical shift values are ≈0.01 and ≈0.05 p.p.m. for ^{1}H and ^{13}C, respectively. The 5' and 5'' protons are not stereospecifically assigned.

[a]Not assigned.

Table S2. Chemical shifts (ppm) of exchangeable proton, protonated nitrogen, and phosphorous resonances.

Residue	H1/H3	N1/N3	HN4/HN6 /HN2	N4/N6/N2	5'-P
G_{27}	[a]n.o.	n.o.	n.o.	n.o.	–
G_{28}	12.70	144.80	[b]n.a.	n.o.	-3.88
G_{29}	12.51	144.76	n.a.	n.o.	-3.69
G_{30}	12.27	144.30	n.a.	n.o.	-3.68
A_{31}	-	-	7.71, 6.51	80.13	-4.01
ψ_{32}	10.31, 13.21	128.59, 158.17	-	-	-3.77
U_{33}	13.21	157.98	-	-	-3.85
G_{34}	10.49	143.16	n.a.	n.o.	-3.86
A_{35}	–	-	[c]6.42	75.71	-3.31
A_{36}	–	-	[c]6.61	76.90	-3.99
A_{37}	-	-	n.a.	n.a.	-3.40
A_{38}	-	-	n.o.	80.05	-3.38
U_{39}	13.86	159.41	-	-	-4.63
C_{40}	-	144.04	6.91, 8.39	95.65	-4.42
C_{41}	-	144.79	6.85, 8.42	95.27	-4.45
C_{42}	-	143.64	6.90, 8.40	95.29	-4.42
C_{43}	–	–	6.97, 8.23	94.14	-4.24

The ^{1}H chemical shifts were referenced relative to DSS at 12 ˚C and pH 6.8 and the ^{15}N chemical shifts set using the spectrometer frequency and the ^{1}H and ^{15}N gyromagnetic ratios. The ^{31}P chemical shifts are reported relative to an external standard of TMP and were recorded at 25 ˚C. The chemical shifts have uncertainties of ≈0.01, ≈0.03, and ≈0.01 ppm for ^{1}H, ^{15}N, and ^{31}P, respectively.

[a]Not observed.

[b]Not assigned.

[c]These resonanc es are broad and weak.