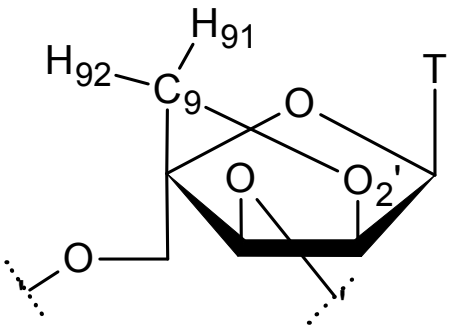
SUPPLEMENTARY MATERIAL

Table S1. RESP derived atomic charges and atom types for α-L-LNA thymine monomer



AMBER atom types differing from standard DNA

atom	atom type
H2'	H1
O2'	OS
C9	CT
H91	H1
H92	H1

atom	charge
O5'	−0.5176
C5'	0.0176
H5'	0.0974
H5"	0.0974
C1'	−0.0166
C2'	0.1170
C3'	0.1525
C4'	0.1439
O4'	−0.3603
H1'	0.1856
H2'	0.1455

atom	charge
H3'	0.0962
O2'	−0.4268
C9	0.0613
H91	0.1025
H92	0.1025
O3'	−0.4991
N1	−0.0320
C2	0.5645
N3	−0.4280
C4	0.5639

atom	charge
C5	0.0251
C6	−0.2612
O2	−0.5504
H3	0.3259
O4	−0.5473
H6	0.2118
C7	−0.2483
H71	0.0876
H72	0.0876
H73	0.0876

Table S2. A selection of chemical shifts (in ppm) for the d(C$^{\alpha L}$T^{L}GA$^{\alpha L}$T^{L}A$^{\alpha L}$T^{L}GC):r(GCAUAUCAG) hybrid. Values are given at 25°C relative to DSS or inorganic phosphorous. [a]

	H6/H8	H1'	H2'	H2''	H3'	H4'	H5/H2/CH_3	H1/H3	^{31}P
C1	7.81	6.28	2.39	2.91	4.85	4.19	5.99		
$^{\alpha L}$T^{L}2	7.41	5.71	5.00		4.82		1.78	–	−3.95
G3	7.70	5.93	2.25	2.67	4.71	4.42		11.98	−4.44
A4	8.14	6.32	2.75	3.05	5.07	4.50	7.73		−3.99
$^{\alpha L}$T^{L}5	6.79	5.41	4.68		4.77		1.55	13.59	−4.18
A6	8.02	6.27	2.55	2.97	4.78	4.45	7.25		−4.22
$^{\alpha L}$T^{L}7	6.81	5.49	4.89		4.77		1.36	13.38	−4.16
G8	7.54	6.04	2.35	2.64	4.52	4.38		12.46	−4.87
C9	7.41	6.15	2.09	2.30	4.44	4.08	5.12		−4.02
G10	7.91	5.55	4.66		4.48	4.22		–	
C11	7.80	5.55	4.66		4.57	4.47	5.29		−3.90
A12	8.01	5.97	4.48		4.64	4.52	7.14		−3.41
U13	7.48	5.43	4.62		4.42	4.45	5.07	12.89	−3.93
A14	8.09	5.93	4.40		4.62	4.54	6.81		−3.65
U15	7.59	5.45	4.35		4.39	4.39	4.85	13.44	−4.23
C16	7.98	5.61	4.52		4.58	4.43	5.67		−4.02
A17	8.00	5.83	4.30		4.70	4.45	6.93		−3.53
G18	7.34	5.77	3.99		4.21	4.47		–	−3.81

[a] The protons in the C2',C4' linker (H6' and H6") have the following chemical shift values:

$^{\alpha L}$T^{L}**2**: 4.36/4.44; $^{\alpha L}$T^{L}**5**: 4.31/4.31; $^{\alpha L}$T^{L}**7**: 4.24/4.24.

Table S3. A selection of helix parameters for the α-L-LNA:RNA hybrid. Values for standard A- and B-type duplexes are included for comparison

	X-disp (Å)	Y-disp (Å)	Tip (°)	Inclin (°)	Prop.Twist (°)		Twist (°)	Roll (°)	Rise (Å)
C1	−3.80	0.54	4.01	13.7	−10.5	C1–T2	23.3	−4.24	2.85
$^{\alpha L}$**T**^{L}2	−3.75	0.21	2.52	15.9	−18.0	T2–G3	37.5	20.98	2.23
G3	−3.81	0.01	7.53	15.8	−20.0	G3–A4	35.5	−3.78	2.70
A4	−3.81	0.29	4.45	14.2	−15.0	A4–T5	26.3	−6.27	3.09
$^{\alpha L}$**T**^{L}5	−3.56	−0.11	2.22	10.1	−11.4	T5–A6	38.4	5.25	2.65
A6	−3.55	−0.01	3.94	9.4	−20.6	A6–T7	29.4	−4.07	3.00
$^{\alpha L}$**T**^{L}7	−3.48	−0.32	3.44	10.1	−19.8	T7–G8	40.4	7.19	2.85
G8	−3.46	−0.29	5.68	10.6	−16.2	G8–C9	31.1	−4.81	2.85
C9	−3.39	−0.18	3.92	11.2	−9.6				
A-type	−5.4	0	0	19.1	−13.7		32.7	0	2.56
B-type	−0.7	0	0	−5.9	−3.7		36.0	0	3.38

Table S4. Backbone torsion angles and sugar puckers (in degrees) in the *in vacuo* structure of the α-L-LNA:RNA hybrid

	α	β	γ	δ	ϵ	ζ	χ	ϕ_{max}	P
C1			47	131	137	−63	−103	28	168
$^{\alpha L}$**T**^{L}2	−172	203	182	−62	174	49	−125	67	14
G3	116	153	160	114	−176	−76	−151	18	123
A4	−78	186	48	133	−166	−89	−105	34	157
$^{\alpha L}$**T**^{L}5	−85	154	172	−63	178	56	−158	61	16
A6	−161	190	80	115	−166	−81	−121	37	129
$^{\alpha L}$**T**^{L}7	−101	163	172	−64	176	65	−150	61	16
G8	−135	183	50	106	−170	−78	−143	22	95
C9	−77	172	57	100			−130	29	88
G10			59	80	−125	−91	−171	38	7
C11	−66	145	65	79	−160	−66	−156	41	9
A12	−78	177	58	79	−164	−62	−159	42	6
U13	−80	176	69	78	−159	−65	−158	40	3
A14	−76	178	57	81	−173	−61	−157	40	6
U15	−79	178	80	82	−159	−62	−152	37	2
C16	−77	172	55	79	−152	−58	−146	39	9
A17	−76	186	36	76	−177	−63	−155	37	16
G18	−76	170	64	72			−160	44	12