Table S1: Chemical shift assignments from solid state NMR spectra of FUS-LC fibrils, Related to Fig. 3. Chemical shifts are in parts per million relative to $NH_{3(l)}$ for ^{15}N and relative to DSS for ^{13}C. Uncertainties in chemical shifts are approximately ±0.25 ppm for ^{15}N and ±0.15 ppm for ^{13}C. Backbone ϕ and ψ torsion angle predictions based on these chemical shifts are from TALOS-N, with error limits representing uncertainties reported by TALOS-N.

residue	chemical shifts (ppm)								predicted torsion angles (°)	
	^{15}N	$^{13}C_\alpha$	^{13}CO	$^{13}C_\beta$	$^{13}C_\gamma$	$^{13}C_\delta$	$^{13}C_\varepsilon$	$^{13}C_\zeta$	ϕ	ψ
S39	118.5	57.6	174.6	65.6						
G40	110.6	47.5								
S44	122.1	56.8	173.2	65.5					-98 ± 14	129 ± 8
T45	122.5	63.1	172.9	71.7	21.9				-114 ± 12	128 ± 6
D46	127.4	50.7	176.3	42.9	180.9				-108 ± 13	124 ± 13
T47	121.8	63.5	173.2	71.5	21.9				-115 ± 16*†	129 ± 12*†
S48	121.4	59.3	176.1	63.4					-76 ± 19	138 ± 14†
G49	114.4	44.5	173.4						77 ± 10	19 ± 18
Y50	104.7	61.7	173.5	37.7	128.2	131.4 133.5	118.8	157.0		
G51	108.3	42.9	172.1							
Q52	110.3	56.4	176.0	27.1	35.2	181.1				
S53	116.4	58.0	173.0	66.5					-120 ± 27	162 ± 10
S54	113.9	56.1	175.0	65.9					-142 ± 13	155 ± 10
N63	126.9	52.4	173.7	39.9	176.4				-96 ± 12	124 ± 9
T64	122.9	61.4	173.6	70.6	21.9				-105 ± 13	131 ± 13
G65	117.2	45.5	171.1						-71 ± 8	161 ± 11
Y66	122.9	57.9	177.5	41.5	130.1	132.0 134.1	118.8	156.9	-62 ± 9	138 ± 10†
G67	114.9	47.1	174.0						80 ± 9*	9 ± 15*†
T68	115.0	61.4	172.0	70.1					-105 ± 20	136 ± 9
Q69	126.8	53.9	173.6	34.2	34.8	177.2			-127 ± 13	146 ± 12
S70	120.5	54.8	173.7	64.2					-103 ± 12	140 ± 11
T71		60.7		70.1	20.5				-67 ± 9	146 ± 11
P72	132.9	62.3	176.6	32.3	28.5	47.2			-65 ± 9	145 ± 10
Q73	121.8	54.7	175.3	33.4	34.9				-118 ± 23	158 ± 14
G74	110.2	46.2	173.9							
Y75	126.6	59.9	177.6	39.7	129.1	131.2 133.5	117.6 118.6	157.0		
G76	103.3	45.7	175.8							
S77	119.1	58.6	174.7	65.2						
T78	123.4	62.6	173.1	69.9	19.8				-107 ± 14	133 ± 9
G79	116.4	43.6	172.1							
G80	109.9	43.6	171.6							
Y81	122.0	61.6	177.1	39.6	131.1	131.7 131.7	117.4 117.7	157.7	-62 ± 6*	141 ± 8*†
G82	110.1	48.7	174.9							
S83	109.7	57.5	172.5	66.0						
S84	116.5	55.9	175.0	67.6					-125 ± 14	149 ± 11†
Q85	128.4	54.7	172.3	33.4	34.1	180.3			-134 ± 12†	150 ± 10†
S86	120.9	56.0	174.0	67.0					-131 ± 8	152 ± 8
S87	113.0	58.9	172.9	65.2					-143 ± 29	155 ± 12
Q88	120.2	56.4	176.4	31.9	34.1	179.0				

S89	117.1	56.6	173.4	66.0					-128 ± 14	148 ± 12
S90	118.2	54.6	173.1	64.4					-116 ± 16	143 ± 18
Y91	123.7	55.2	175.8	39.2	129.1	132.6 133.4	117.1	157.8		
G92	100.3	46.6	174.4							
Q93	116.2	58.3	175.6	28.7	36	177.5			-66 ± 8*	139 ± 12*
Q94	117.5	54.8	175.6	34.2		177.7			-128 ± 13	147 ± 12
S95	121.7	56.2	173.8	65.2					-101 ± 24	151 ± 15

*Predictions classified as "generous" by TALOS-N. All other predictions are "strong".

†Predictions not used as restraints in structure calculations, due to conflicts with ^{15}N-BARE data.

Table S2. Chemical shifts of unassigned signals in 3D solid state NMR spectra of FUS-LC fibrils, Related to Fig. 3. Possible assignments are listed when a signal was assigned to residues 1-112 in at least one of 50 mcassign2b runs with a connection to a neighboring residue.

3D spectrum	chemical shifts (ppm)					residue type	possible assignments
	^{15}N	$^{13}C_{\alpha}$	^{13}CO	$^{13}C_{\beta}$	$^{13}C_{\gamma}$		
NCACX	108.2	45.2				G	111
	111.5	58.7	171.7	69.1		S	-
	114.1	59.3	171.7	65.9		S	26, 30
	114.5	55.8	171.9	67.1		S	26, 30, 61
	117.2	56.2	171.6	65.0		S	42, 57
	120.5	54.7	173.6	28.5	35.0	Q	36, 62
	121.0	56.4	176.4	31.4	34.2	Q	27, 31, 43
	121.5	54.6	174.2	32.9	35.0	Q	27, 31
	123.7	54.3	173.0	32.0	33.8	Q	31, 36, 103
	124.3	57.0	173.8			Q, S, Y, N, or D	-
	125.4	54.9	177.7	34.0		Q	-
NCOCX	114.1	55.2	175.5			Q, S, Y, N, or D	41, 55
	114.5	58.8	171.8	69.1		S	110, 112
	117.2	54.9	174.8			Q, S, Y, N, or D	55, 56
	120.2	56.2	172.9			Q, S, Y, N, or D	27, 35, 61
	121.0	55.7	172.0	67.0		S	26, 30, 42, 61
	121.9	59.3	171.8	66.0		S	26, 30
	123.7	57.0	173.9			Q, S, Y, N, or D	8, 36, 102
	125.2	59.9	171.4	65.5		Q, S, Y, N, or D	-
	126.8	54.3	173.1	32.5	33.7	Q	-
	127.8	56.2	171.5	64.9		S	112
	107.6	48.9	177.2			G	-
CONCA	111.5	58.6	175.5			Q, S, Y, N, or D	112
	112.4	55.3	171.6			Q, S, Y, N, or D	41
	114.5	59.2	176.0			Q, S, Y, N, or D	26, 30
	114.6	55.7	172.3			Q, S, Y, N, or D	26, 30
	117.8	55.9	174.9			Q, S, Y, N, or D	27, 35, 102
	121.4	54.6	172.0			Q, S, Y, N, or D	27, 31
	122.4	56.6	174.0			Q, S, Y, N, or D	35, 37, 61, 103
	123.3	58.4	174.5			Q, S, Y, N, or D	-
	123.6	54.3	174.0			Q, S, Y, N, or D	9, 31, 37, 103
	125.0	54.9	171.6			Q, S, Y, N, or D	36, 56, 103
	126.8	55.1	171.6			Q, S, Y, N, or D	-
	112.7	55.7	58.2	173.0		Q, S, Y, N, or D	-

Table S3. Summary of NMR measurement conditions, Related to Fig. 2.[1]

Sample	Spectrum	NMR parameters	Total time	Processing
U-FUS-LC, 10 mg	2D CC	B_0 = 17.5 T; ν_{MAS} = 17.0 kHz; na = 240; τ_{pd} = 2.5 s; t_{1max} = 8.6 ms; t_{1inc} = 24 μs; τ_{dwell} = 15 μs; τ_{acq} = 15.4 ms; τ_{HC} =1.5 ms; τ_{DARR} = 25 ms; ν_{1H} = 85 kHz	120 h	GB = 75 Hz in t_1; GB = 75 Hz in t_2
U-FUS-LC, 10 mg	2D NCA	B_0 = 21.1 T; ν_{MAS} = 11 kHz; na = 64; τ_{pd} = 2.0 s; t_{1max} = 12.7 ms; t_{1inc} = 201.6 μs; τ_{dwell} = 10 μs; τ_{acq} = 20.5 ms; τ_{HC} = 1 ms; τ_{NC} = 4 ms; ν_{1H} = 85 kHz; ν_{1C} = 42 kHz, ν_{1N} = 31 kHz, and ν_{0C} = 53 ppm during τ_{NC}	5 h	GB = 10 Hz in t_1; GB = 50 Hz in t_2
U-FUS-LC, 10 mg	2D HC INEPT	B_0 = 14.1 T; ν_{MAS}= 12 kHz; na = 32; τ_{pd} = 2.0 s; t_{1max} = 10 ms; t_{1inc} = 50 μs; τ_{dwell} = 15 μs; τ_{acq} = 15.4 ms; ν_{1H} = 13 kHz	7 h	GB = 10 Hz in t_1: GB = 50 Hz in t_2
U-FUS-LC, 10 mg	2D HN INEPT	B_0 = 17.5 T; ν_{MAS}=17.0 kHz; na = 64; τ_{pd} = 2.0 s; t_{1max} = 14 ms; t_{1inc} = 200 μs; τ_{dwell} = 15 μs; τ_{acq} = 30.7 ms; ν_{1H} = 38 kHz	5 h	GB = 0 Hz in t_1: GB = 0 Hz in t_2
U-FUS-LC, 10 mg	3D NCACX	B_0 = 21.1 T; ν_{MAS} = 13.8 kHz; na = 8; τ_{pd} = 2.0 s; t_{1max} = 12 ms; t_{1inc} = 222 μs; t_{2max} = 6.8 ms; t_{2inc} = 109 μs; τ_{dwell} = 5 μs; τ_{acq} = 10.2 ms; τ_{HN} = 1 ms; τ_{NC} = 4 ms; τ_{DARR} = 50 ms; ν_{1H} = 85 kHz; ν_{1C} = 22 kHz, ν_{1N} = 36 kHz, and ν_{0C} = 53 ppm during τ_{NC}	60 h	GB = 15 Hz in t_1; GB = 25 Hz in t_2; GB = 25 Hz in t_3
U-FUS-LC, 10 mg	3D NCOCX	B_0 = 21.1 T; ν_{MAS} = 13.8 kHz; na = 16; τ_{pd} = 2.0 s; t_{1max} = 12 ms; t_{1inc} = 222 μs; t_{2max} =9.1 ms; t_{2inc} = 222 μs; τ_{dwell} = 5 μs; τ_{acq} = 10.2 ms; τ_{HN} = 1 ms; τ_{NC} = 4 ms; τ_{DARR} = 50 ms; ν_{1H} = 83 kHz; ν_{1C} = 52 kHz, ν_{1N} = 39 kHz, and ν_{0C} = 175 ppm during τ_{NC}	79 h	GB = 15 Hz in t_1; GB = 25 Hz in t_2; GB = 25 Hz in t_3
U-FUS-LC, 10 mg	3D CONCA	B_0 = 14.1 T; ν_{MAS} = 12 kHz; na = 16; τ_{pd} = 2.0 s; t_{1max} = 5.9 ms; t_{1inc} = 196 μs; t_{2max} = 11.8 ms; t_{2inc} = 196 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{CN} = 4 ms; τ_{NC} = 4 ms; ν_{1H} = 70 kHz; ν_{1C} = 29 kHz, ν_{1N} = 17 kHz, and ν_{0C} = 168 ppm during τ_{CN}; ν_{1C} = 29 kHz, ν_{1N} = 17 kHz, and ν_{0C} = 60 ppm during τ_{NC}	68 h	GB = 25 Hz in t_1; GB = 15 Hz in t_2; GB = 25 Hz in t_3
U-FUS-LC, 10 mg	3D CANCX	B_0 = 14.1 T; ν_{MAS} = 12 kHz; na = 256; τ_{pd} = 2.0 s; t_{1max} = 4.0 ms; t_{1inc} = 165.2 μs; t_{2max}= 6.6 ms; t_{2inc} = 165.2 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{CN} = 4 ms; τ_{NC} = 4 ms; τ_{DARR} = 60 ms; ν_{1H} = 85 kHz; ν_{1C} = 26 kHz, ν_{1N} = 14 kHz, and ν_{0C} = 56 ppm during τ_{CN}; ν_{1C} = 16 kHz, ν_{1N} = 4 kHz, and ν_{0C} = 175 ppm during τ_{NC}	578 h	GB = 50 Hz in t_1; GB = 30 Hz in t_2; GB = 50 Hz in t_3
N112-FUS-LC, 6 mg	1D HC CP	B_0 = 14.1 T; ν_{MAS} =13.6 kHz; na = 32; τ_{pd} = 3.0 s, τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms, τ_{HC} =1.5 ms, ν_{1H} = 93 kHz	0.03 h	GB = 80 Hz
N112-FUS-LC, 6 mg	1D HC INEPT	B_0 = 14.1 T; ν_{MAS}=13.6 kHz; na = 128; τ_{pd}= 2.0 s; τ_{dwell} = 15 μs; τ_{acq} = 30.7 ms; ν_{1H} = 16 kHz	0.07 h	GB = 20 Hz
N112-FUS-LC, 6 mg	2D CC	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 32; τ_{pd} = 3.0 s; t_{1max} = 5.5 ms; t_{1inc} = 21.6 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{DARR} = 50 ms; ν_{1H} = 93 kHz	14 h	GB = 75 Hz in t_1: GB = 75 Hz in t_2
N112-FUS-LC, 6 mg	2D NCA	B_0 = 14.1 T; ν_{MAS}=13.6 kHz; na = 384; τ_{pd} = 2.2 s; t_{1max} = 12.4 ms; t_{1inc} = 129.6 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HN} = 1.5 ms; τ_{NC} = 4 ms; ν_{1H} = 93 kHz; ν_{1C} = 34 kHz, ν_{1N} = 21 kHz, and ν_{0C} = 55 ppm during τ_{NC}	45 h	GB = 0 Hz in t_1: GB = 75 Hz in t_2
N112-FUS-LC, 6 mg	3D NCACX	B_0 = 17.5 T; ν_{MAS} = 17 kHz; na = 32; τ_{pd} = 1.5 s; t_{1max} = 6.9 ms; t_{1inc} = 172.8 μs; t_{2max} = 5.2 ms; t_{2inc} = 64.8 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{NC} = 5 ms; ; τ_{DARR} = 50 ms; ν_{1H} = 93 kHz; ν_{1C} = 43 kHz, ν_{1N} = 26 kHz, and ν_{0C} = 51 ppm during τ_{NC};	170 h	GB = 10 Hz in t_1; GB = 10 Hz in t_2; GB = 100 Hz in t_3
N112-FUS-LC, 6 mg	3D CONCA	B_0 = 17.5 T; ν_{MAS} = 17 kHz; na = 128; τ_{pd} = 2.0 s; t_{1max} = 4.4 ms; t_{1inc} = 259.2 μs; t_{2max} = 6.9 ms; t_{2inc} = 172.8 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{CN} = 4 ms; τ_{NC} = 5 ms; ν_{1H} = 93 kHz; ν_{1C} = 43 kHz, ν_{1N} = 26 kHz, and ν_{0C} = 175 ppm during τ_{CN}; ν_{1C} = 43 kHz, ν_{1N} = 26 kHz, and ν_{0C} = 51 ppm during τ_{NC}	192 h	GB = 10 Hz in t_1; GB = 10 Hz in t_2; GB = 100 Hz in t_3
C112-FUS-LC, 4 mg	1D HC CP	B_0 = 14.1 T; ν_{MAS}=13.6 kHz; na = 128; τ_{pd} = 2.0 s; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 15 ms, ν_{1H} = 90 kHz	0.07 h	GB = 80 Hz
C112-FUS-LC, 4 mg	1D HC INEPT	B_0 = 14.1 T; ν_{MAS}=13.6 kHz; na =128; τ_{pd}= 2.0 s; τ_{dwell} = 15 μs; τ_{acq} = 30.7 ms; ν_{1H} = 16 kHz	0.07 h	GB = 20 Hz
C112-FUS-LC, 4 mg	2D CC	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 192; τ_{pd} = 2.2 s; t_{1max} = 4 ms; t_{1inc} = 22.4 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{DARR} = 50 ms; ν_{1H} = 78 kHz	42 h	GB = 75 Hz in t_1: GB = 75 Hz in t_2
C112-FUS-LC, 4 mg	2D NCA	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 2880; τ_{pd} = 2.2 s; t_{1max} = 8.6 ms; t_{1inc} = 134.4 μs; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HN} = 1.5 ms; τ_{NC} = 4 ms; ν_{1H} = 78 kHz; ν_{1C} = 34 kHz, ν_{1N} = 21 kHz, and ν_{0C} = 55 ppm during τ_{NC}	225 h	GB = 50 Hz in t_1: GB = 75 Hz in t_2
N60-FUS-LC, 4 mg	1D HC CP	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 1024; τ_{pd} = 2.0 s; τ_{dwell} = 15 μs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; ν_{1H} = 90 kHz	0.6 h	GB = 80 Hz

N60-FUS-LC, 4 mg	1D HC INEPT	B_0 = 14.1 T; ν_{MAS}=13.6 kHz; na = 1024; τ_{pd} = 1.5 s; τ_{dwell} = 15 µs; τ_{acq} = 30.7 ms; ν_{1H} = 13 kHz	0.4 h	GB = 20 Hz
N60-FUS-LC, 4 mg	2D CC	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 128; τ_{pd} = 2.0 s; t_{1max} = 5.4 ms; t_{1inc} = 21.2 µs; τ_{dw} = 15 µs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{DARR} = 50 ms; ν_{1H} = 93 kHz	36 h	GB = 0 Hz in t_1: GB = 75 Hz in t_2
N60-FUS-LC, 4 mg	2D NCA	B_0 = 17.5 T; ν_{MAS} = 17.0 kHz; na = 512; τ_{pd} = 2.0 s; t_{1max} = 5.4 ms; t_{1inc} = 84 µs; τ_{dwell} = 15 µs; τ_{acq} = 7.7 ms; τ_{HN} = 1.5 ms; τ_{NC} = 4 ms; ν_{1H} = 83 kHz; ν_{1C} = 24 kHz, ν_{1N} = 7 kHz, and ν_{0C} = 53 ppm during τ_{NC}	36 h	GB = 55 Hz in t_1: GB = 75 Hz in t_2
C60-FUS-LC, 3 mg	1D HC CP	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 1024; τ_{pd} = 2.0 s; τ_{dwell} = 15 µs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; ν_{1H} = 90 kHz	0.6 h	GB = 80 Hz
C60-FUS-LC, 3 mg	1D HC INEPT	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 1024; τ_{pd} = 1.5 s; τ_{dwell} = 15 µs; τ_{acq} = 30.7 ms; ν_{1H} = 13 kHz	0.4 h	GB = 20 Hz
C60-FUS-LC, 3 mg	2D CC	B_0 = 14.1 T; ν_{MAS} = 13.6 kHz; na = 192; τ_{pd} = 2.0 s; t_{1max} = 5.4 ms; t_{1inc} = 21.2 µs; τ_{dwell} = 15 µs; τ_{acq} = 7.7 ms; τ_{HC} = 1.5 ms; τ_{DARR} = 50 ms; ν_{1H} = 90 kHz	55 h	GB = 0 Hz in t_1: GB = 75 Hz in t_2
C60-FUS-LC, 3 mg	2D NCA	B_0 = 17.5 T; ν_{MAS} = 17.0 kHz; na = 896; τ_{pd} = 2.0 s; t_{1max} = 5.4 ms; t_{1inc} = 84 µs; τ_{dwell} = 15 µs; τ_{acq} = 7.7 ms; τ_{HN} = 1.5 ms; τ_{NC} = 4 ms; ν_{1H} = 83 kHz; ν_{1C} = 24 kHz, ν_{1N} = 7 kHz, and ν_{0C} = 53 ppm during τ_{NC}	64 h	GB = 55 Hz in t_1: GB = 75 Hz in t_2
1-^{13}C-Tyr-FUS-LC, 5 mg	PITHIRDS-CT	B_0 = 9.4 T; ν_{MAS} = 20.0 kHz; na = 256; τ_{pd} = 4 s; ν_{1H} = 100 kHz during 38.4 ms constant-time recoupling period; τ_{dwell} = 20 µs; τ_{acq} = 41.0 ms; ν_{1H} = 70 kHz with pulsed spin-locking of ^{13}C during τ_{acq}	2.6 h	GB = 20 Hz
1-^{13}C-Thr-FUS-LC, 5 mg	PITHIRDS-CT	B_0 = 9.4 T; ν_{MAS} = 20.0 kHz; na = 1216; τ_{pd} = 4 s; ν_{1H} = 100 kHz during 38.4 ms constant-time recoupling period; τ_{dwell} = 20 µs; τ_{acq} = 41.0 ms; ν_{1H} = 70 kHz with pulsed spin-locking of ^{13}C during τ_{acq}	12.5 h	GB = 20 Hz
U-FUS-LC, 10 mg	^{15}N-BARE	B_0 = 17.5 T; ν_{MAS} = 17 kHz; na = 384; τ_{pd} = 2.0 s; t_{1max} = 8.9 ms; t_{1inc} = 120 µs; τ_{dwell} = 15 µs; τ_{acq} = 5.5 ms; τ_{HN} = 1.5 ms; $\tau_{NCfsTEDOR}$ = 0.7 ms; ν_{1H} = 85 kHz during acquisition and t_1 evolution; ν_{0C} = 56 ppm during τ_{NC}; τ_{SD} = 30 ms; 28.2 ms constant-time recoupling period; ν_{NBARE} = 25.5 kHz; 6 periods of recoupling with 5.6 ms increment between blocks; ν_{1H} = 100 kHz during constant time recoupling block	32 h per recoupling increment	GB = 125 Hz in t_1: GB = 60 Hz in t_2
2--Glyc-FUS-LC, 10 mg	^{15}N-BARE	B_0 = 17.5 T; ν_{MAS} = 12 kHz; na = 192; τ_{pd} = 2.3 s; t_{1max} = 7.0 ms; t_{1inc} = 100.8 µs; τ_{dwell} = 15 µs; τ_{acq} = 5.5 ms; τ_{HN} = 1.5 ms; τ_{NC} = 4 ms; ν_{1H} = 85 kHz; ν_{1C} = 28 kHz, ν_{1N} = 16 kHz; and ν_{0C} = 53 ppm during τ_{NC}; 28 ms constant-time recoupling period; ν_{NBARE} = 18 kHz; 8 periods of recoupling with 4 ms increment between blocks	17 h per recoupling increment	GB = 0 Hz in t_1: GB = 0 Hz in t_2
2-Glyc-FUS-LC, 10 mg	3D NCACX	B_0 = 21.1 T; ν_{MAS} = 13.95 kHz; na = 16; τ_{pd} = 2.0 s; t_{1max} = 8.8 ms; t_{1inc} = 180 µs; t_{2max} = 6.1 ms; t_{2inc} = 105 µs; τ_{dwell} = 5 µs; τ_{acq} = 12.0 ms; τ_{HN} = 0.8 ms; τ_{NC} = 6 ms; τ_{DARR} = 400 ms; ν_{1H} = 83.3 kHz; ν_{1C} = 22 kHz, ν_{1N} = 36 kHz, and ν_{0C} = 56 ppm during τ_{NC}	101 h	GB = 15 Hz in t_1; GB = 25 Hz in t_2; GB = 25 Hz in t_3
2-Glyc-FUS-LC, 10 mg	2D CC	B_0 = 17.5 T; ν_{MAS} = 11.7 kHz; na = 80; τ_{pd} = 1.5 s; t_{1max} = 7.0 ms; t_{1inc} = 22.4 µs; τ_{dwell} = 15 µs; τ_{acq} = 15.4 ms; τ_{HC} = 1.2 ms; τ_{DARR} = 300 ms; ν_{1H} = 85 kHz	21 h	GB = 50 Hz in t_1: GB = 50 Hz in t_2
1,3-Glyc-FUS-LC, 10 mg	3D NCOCX	B_0 = 21.1 T; ν_{MAS} = 13.8 kHz; na = 24; τ_{pd} = 2.0 s; t_{1max} = 9.1 ms; t_{1inc} = 190 µs; t_{2max} = 7.5 ms; t_{2inc} = 166 µs; τ_{dwell} = 5 µs; τ_{acq} = 12.0 ms; τ_{HN} = 0.6 ms; τ_{NC} = 5 ms; τ_{DARR} = 400 ms; ν_{1H} = 83.3 kHz; ν_{1C} = 35 kHz, ν_{1N} = 48 kHz, and ν_{0C} = 176 ppm during τ_{NC}	115 h	GB = 15 Hz in t_1; GB = 25 Hz in t_2; GB = 25 Hz in t_3
1,3-Glyc-FUS-LC, 10 mg	2D CC	B_0 = 17.5 T; ν_{MAS} = 12.0 kHz; na = 48; τ_{pd} = 1.5 s; t_{1max} = 9.0 ms; t_{1inc} = 22.4 µs; τ_{dwell} = 15 µs; τ_{acq} = 15.4 ms; τ_{HC} = 1.2 ms; τ_{DARR} = 400 ms; ν_{1H} = 85 kHz	16 h	GB = 50 Hz in t_1: GB = 50 Hz in t_2

[1]B_0 = magnetic field; ν_{MAS} = MAS frequency; ν_{1H} = ^{1}H radio-frequency field amplitude for decoupling; ν_{1X} = radio-frequency field amplitude for cross-polarization, X= N or C (^{15}N or ^{13}C); ν_{0C} = ^{13}C radio-frequency carrier frequency; na = number of scans per free-induction-decay; τ_{pd} = delay between scans; t_{1max} = maximum t_1 value; t_{1inc} = t_1 increment; t_{2max} = maximum t_2 value; t_{2inc} = t_2 increment; τ_{dwell} = digitization dwell time in free-induction-decay; τ_{acq} = free-induction-decay acquisition time; τ_{XY} = cross-polarization period, where X and Y are H, N, or C (^{1}H, ^{15}N, or ^{13}C); τ_{DARR} = DARR mixing time; GB = pure Gaussian line-broadening before Fourier transformation.

Table S4. Summary of structural restraints in Xplor-NIH calculations, Related to Fig. 4.

Xplor-NIH potential term	Experimental basis	Restraints (per monomer)	Round 1 scale factor	Round 2 scale factor	Lowest model energy	Restraint range[1]	Average violation[2]
PosDiffPot (noncrystallographic symmetry)	single set of chemical shifts	-	100	100	18.26	-	-
DistSymmPot (translational symmetry)	single set of chemical shifts, MPL data, cross-β structure	-	10000	10000	0.15	-	-
C-C RDC (intermolecular alignment with z-axis)	cross-β structure	11	100	100	7.86	-	-
C-O RDC (alignment of β-sheet carbonyl groups with z-axis)	cross-β structure	23	0.0	0.01-100	0.23	-	-
CDIH (backbone conformation)	TALOS-N predictions	51	5000	5000	1.67	δ ± (2ε+15°)	0.37 ± 0.33°
TorsionInterpolPot (backbone conformation)	^{15}N-BARE data	33	0.001-5.0	0.2-2.0	307.86	-	-
NOE (intermolecular distance and alignment)	^{13}C PITHIRDS-CT data	11	100	100	98.07	4.75 ± 0.05 Å	0.115 ± 0.050 Å
NOE (long-range contacts)	inter-residue crosspeaks with unique assignments	37	1-100	100	38.37	5.0 ± 3.0 Å	0.18 ± 0.13 Å
NOE (long-range contacts)	inter-residue crosspeaks with partially ambiguous assignments	16	0.01-100	100	15.33	5.0 ± 3.0 Å	0.22 ± 0.10 Å
NOE (long-range contacts)	inter-residue crosspeaks with fully ambiguous assignments	36	0.0001-100	100	9.41	5.0 ± 3.0 Å	0.13 ± 0.08 Å
NOE (backbone conformation)	^{15}N-BARE data	68	10-1000	1000	1.00	site-dependent	0.018 ± 0.016 Å
RepelPot	standard atomic radii	-	0.004-4.0	0.004-4.0	101.59	-	-
TorsionDB	low-energy sidechain conformations	-	0.0	0.002-0.4	3828.97	-	-
BOND	standard bond lengths	-	default	default	22.33	default	0.002 ± 0.002 Å
ANGL	standard bond angles	-	0.4-1.0	0.4-1.0	347.36	default	0.19 ± 0.30 Å
IMPR	standard bond geometry	-	0.4-1.0	0.4-1.0	62.60	default	0.13 ± 0.21°

[1]For CDIH potentials, δ and ε are the average prediction and uncertainty from TALOS-N, respectively.

[2]Average violations are the deviations outside the specified ranges, averaged only over distances or angles that exceed the specified ranges for the 20 structures in PDB 5W3N. Uncertainties are standard deviations.

Table S5. Summaries of statistics, Related to Figs. 4 and 6.

<table>
<tr><td colspan="4">FUS-LC structure calculations</td></tr>
<tr><td colspan="3">Short range inter-residue distances
($1 < | i - j | < 3$, for residues i and j)</td><td>338</td></tr>
<tr><td colspan="3">Long range inter-residue distances ($| i - j | \geq 3$)</td><td>89</td></tr>
<tr><td colspan="3">backbone torsion angle restraints (from TALOS-N)</td><td>51</td></tr>
<tr><td colspan="3">backbone conformational restraints (from ^{15}N-BARE data)</td><td>33</td></tr>
<tr><td colspan="3">MolProbity Clashscore[1]</td><td>1</td></tr>
<tr><td colspan="3">MolProbity Ramachandran outliers[2]</td><td>9.7</td></tr>
<tr><td colspan="3">MolProbity sidechain conformer outliers[2]</td><td>14.5</td></tr>
<tr><td colspan="3">MolProbity standard geometry outliers[1]</td><td>0</td></tr>
<tr><td colspan="3">All heavy atom RMSD
(residues 44-54 and 63-95)</td><td>1.4 Å</td></tr>
<tr><td colspan="3">C_α RMSD
(residues 44-54 and 63-95)</td><td>1.1 Å</td></tr>
<tr><td colspan="4"></td></tr>
<tr><td colspan="4">Effects of DNA-PK phosphorylation</td></tr>
<tr><td colspan="2">Hydrogel binding (threshold ≈ 0.3)</td><td colspan="2">Liquid-like droplet melting
(threshold ≈ 0.2)</td></tr>
<tr><td>site</td><td>location[3]</td><td>site</td><td>location</td></tr>
<tr><td>T19</td><td>NC</td><td>T19</td><td>NC</td></tr>
<tr><td>S30</td><td>NC</td><td>S42</td><td>C</td></tr>
<tr><td>S42</td><td>C</td><td>S54</td><td>C</td></tr>
<tr><td>S54</td><td>C</td><td>T68</td><td>C</td></tr>
<tr><td>S61</td><td>C</td><td>S84</td><td>C</td></tr>
<tr><td>T68</td><td>C</td><td>S87</td><td>C</td></tr>
<tr><td>S84</td><td>C</td><td></td><td></td></tr>
<tr><td>S87</td><td>C</td><td></td><td></td></tr>
<tr><td>success rate</td><td>6/8</td><td></td><td>5/6</td></tr>
<tr><td>probability if random[4]</td><td>0.0047</td><td></td><td>0.0053</td></tr>
</table>

[1]Reported as the number of clashes or outliers per 1000 atoms in the 20 structures in PDB 5W3N.
[2]Reported as the percentile score with respect to all structures.
[3]C = core-forming segment; NC = non-core-forming segments.
[4]Hypergeometric statistics, based on 214 total residues, with 57 C residues and 157 NC residues.