Supporting Information for

A Timesaving Strategy for MAS NMR Spectroscopy by Combining Non-Uniform Sampling and Paramagnetic Relaxation Assisted Condensed Data Collection

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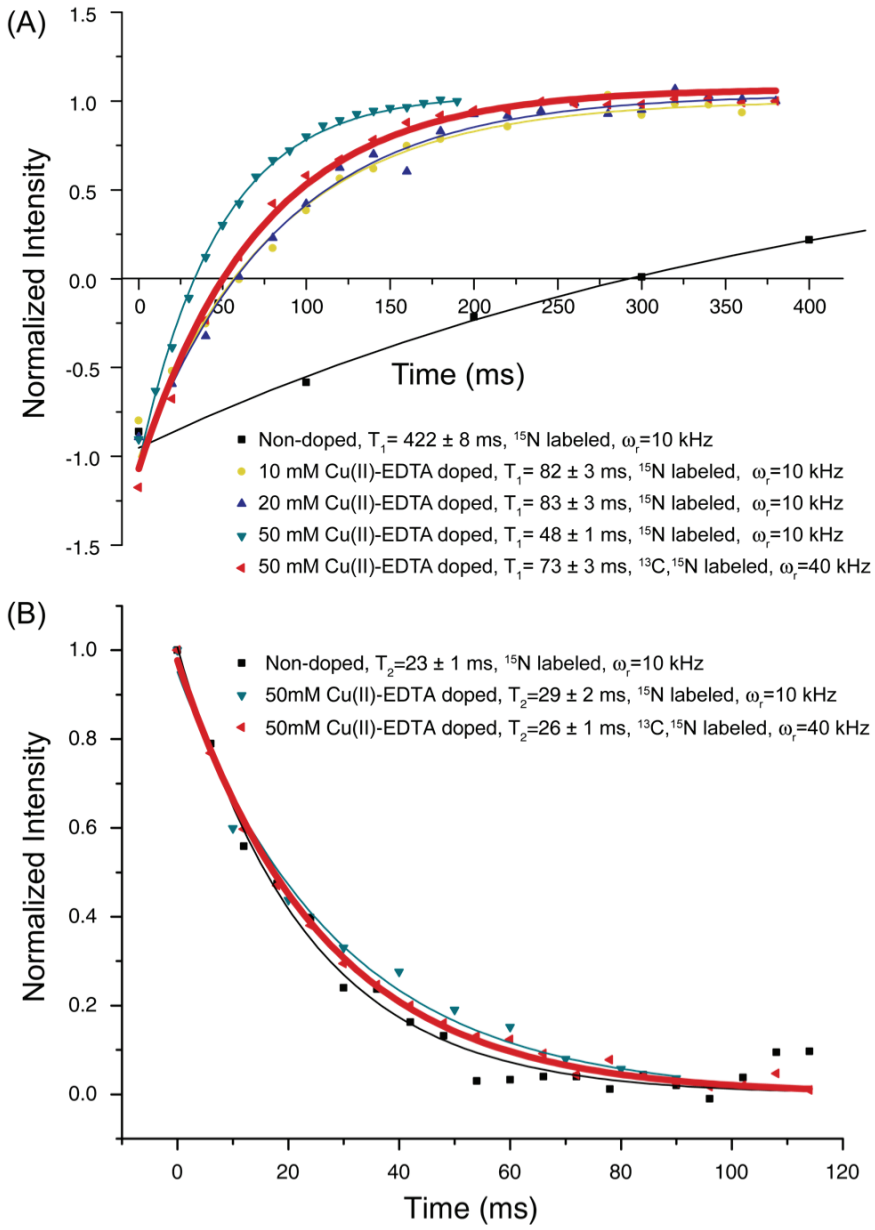

Figure S1. (A) ^{1}H T_1 curves of U-^{13}C,^{15}N-labeled LC8 doped with 50 mM Cu(II)-EDTA (red trace, bold line), and of ^{15}N labeled LC8, neat ("0 mM") (black trace), and Cu(II)-EDTA doped at 10 mM (yellow), 20 mM (blue trace), and 50 mM (dark cyan trace). (B) ^{15}N T_2 curves of U-^{13}C,^{15}N-labeled LC8 doped with 50 mM Cu(II)-EDTA (red trace, bold line), and of ^{15}N labeled LC8, neat ("0 mM") (black trace) and 50 mM (dark cyan trace). Experiments were carried out at the MAS frequency of 10 kHz for ^{15}N labeled LC8 and 40 kHz for U-^{13}C,^{15}N-labeled LC8. Points represent normalized intensities for peaks displaying the largest magnitude in 1D spectra, and are fitted to the following equations: $I = I_0\ [1-2exp(-t/T_1)]$ and $I = I_0\ exp(-t/T_2)$ to extract ^{1}H T_1 and ^{15}N T_2, respectively.

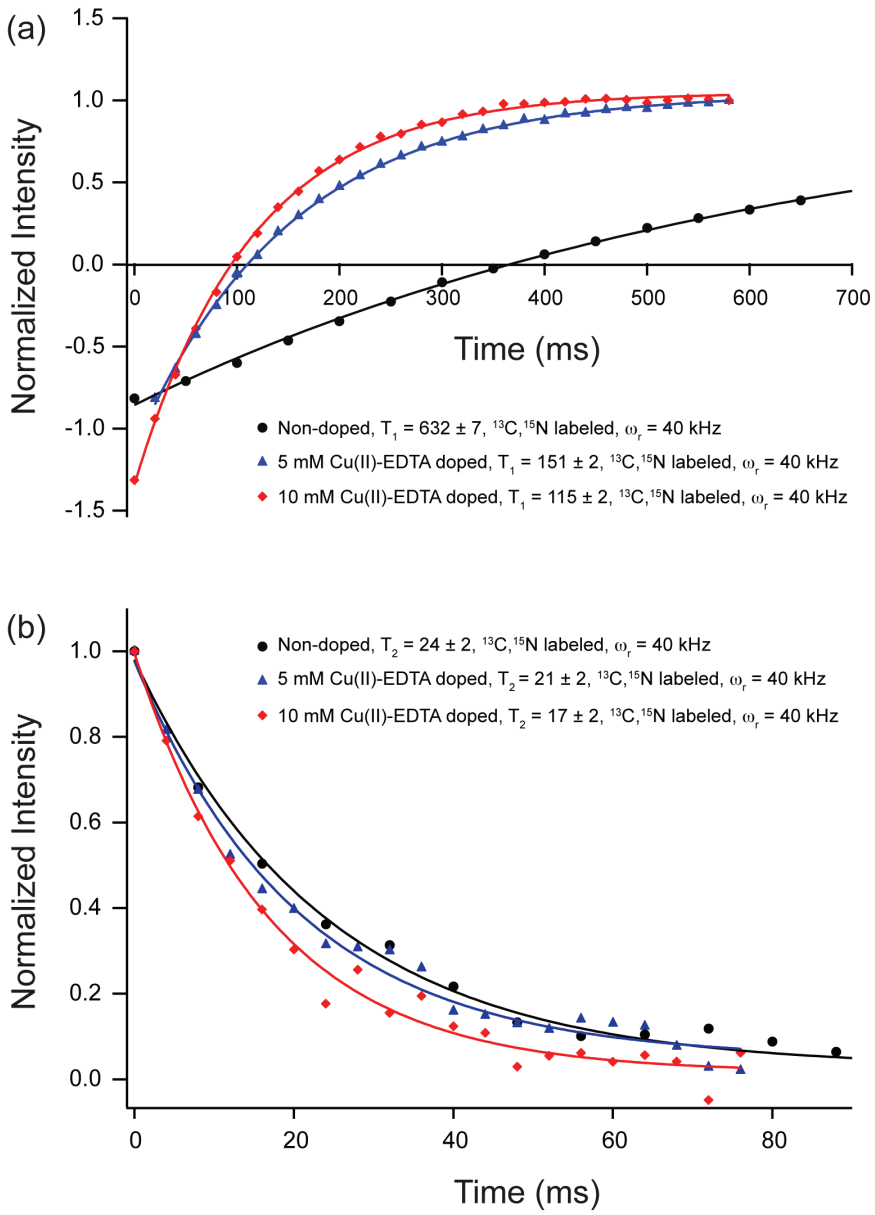

Figure S2. (A) ^{1}H T_1 curves of U-^{13}C,^{15}N-labeled neat LC8 (black trace), and LC8 doped with Cu(II)-EDTA at 5 mM (blue) and 10 mM (red) (B) ^{15}N T_2 curves of U-^{13}C,^{15}N-labeled neat LC8 (black trace), and LC8 doped with Cu(II)-EDTA at 5 mM (blue) and 10 mM (red). Experiments were carried out at the MAS frequency of 40 kHz. Points represent normalized intensities for peaks displaying the largest magnitude in 1D spectra, and are fitted to the following equations: $I = I_0\,[1-2\exp(-t/T_1)]$ and $I = I_0\,\exp(-t/T_2)$ to extract ^{1}H T_1 and ^{15}N T_2, respectively.

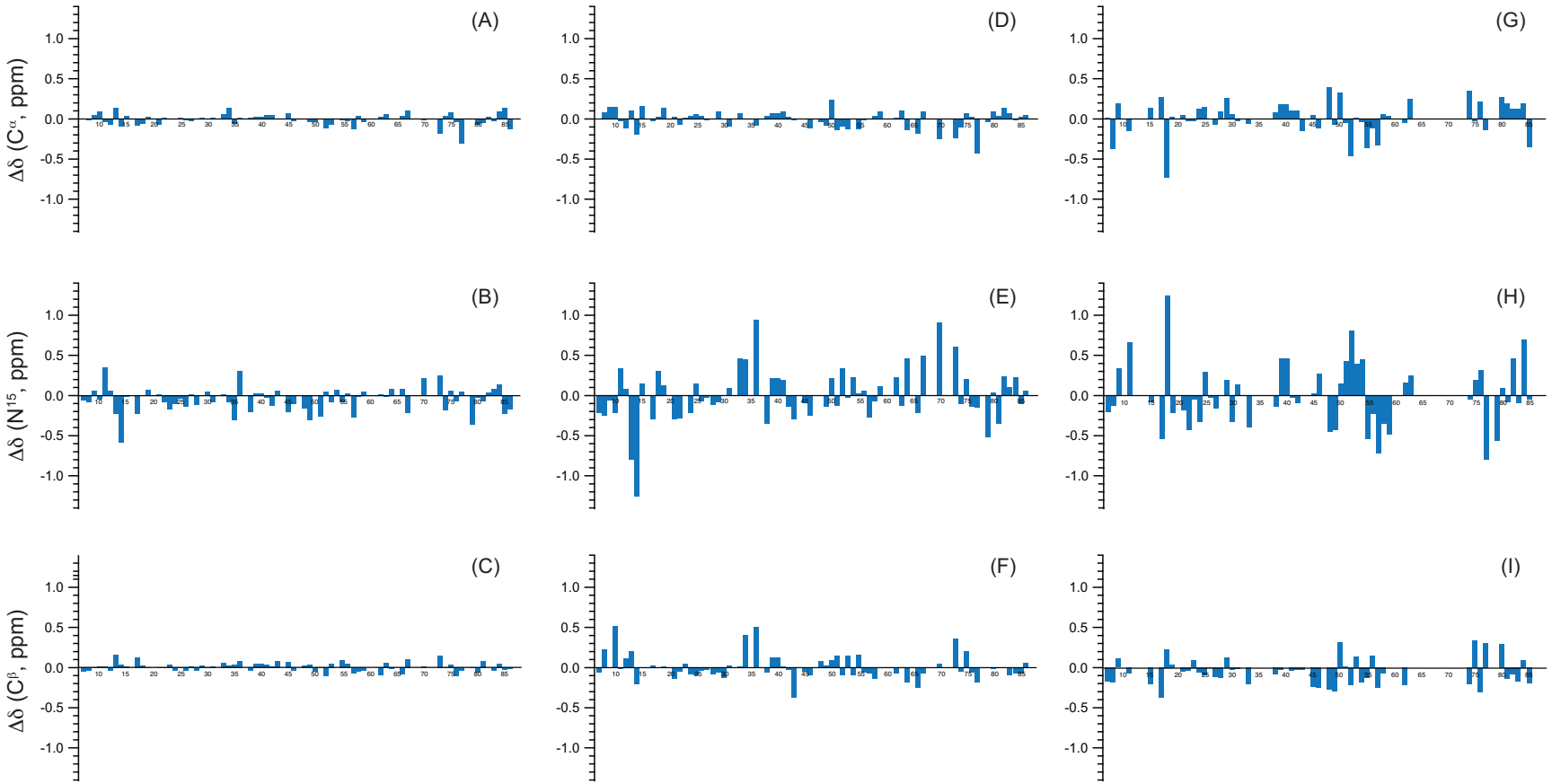

Figure S3. The chemical shift deviations between 3D NCACB spectrum of neat LC8 and 3D NCACB spectra of samples doped with different concentration of Cu(II)-EDTA: 5 mM (left column (A)-(C)), 10 mM (middle column (D)-(F)), 50 mM (right column (G)-(I)).

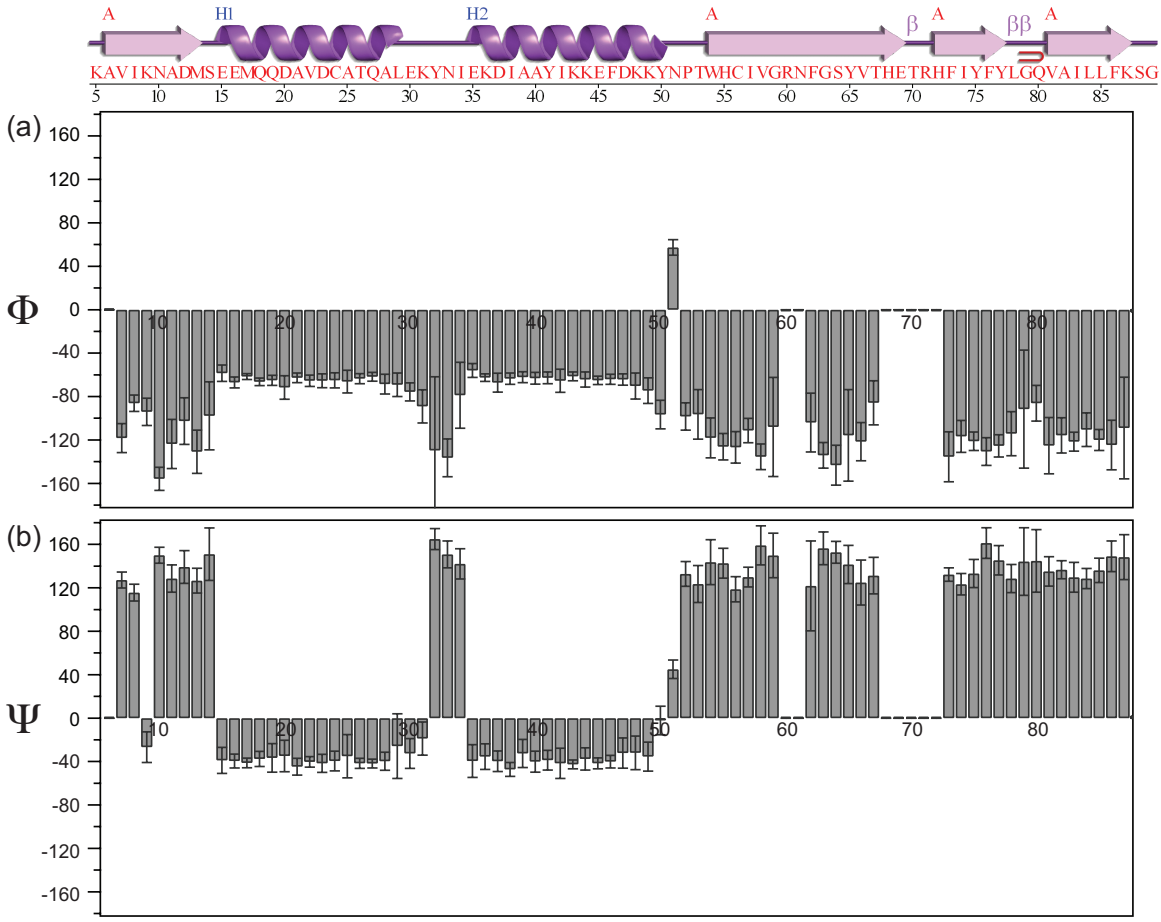

Figure S4. The torsion angles from the NUS-PACC assignments from 5 mM Cu(II)-EDTA LC8 predicted by TALOS+. The predicted secondary structures are highly consistent with X-ray structure and former solution/solid NMR results.

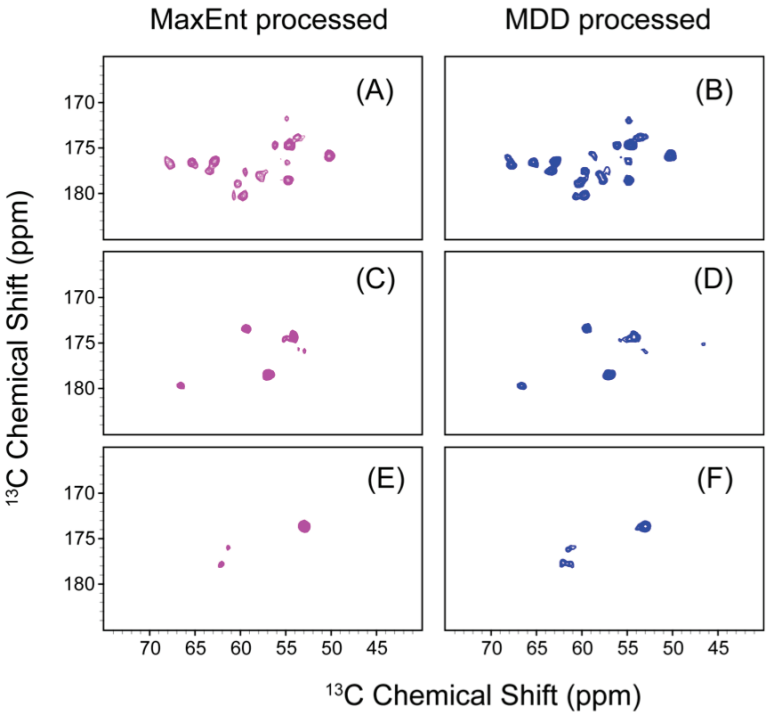

Figure S5. Representative 2D planes of NUS-PACC-NCOCAprocessed with MaxEnt and MDD. The spectra were acquired on a LC8 sample doped with 50 mM Cu(II)-EDTA. (A), (C), (E) are planes for MaxEnt-processed NUS-NCOCA-324 spectrum; (B), (D), (F) are planes for MDD-processed NUS-NCOCA spectrum. Comparison of (A), (C), (E) with (B), (D), (F) indicates that MaxEnt- and MDD- processed spectra are very similar. .

Table S1. Solution chemical shift perturbations observed in the ^{15}N-^{1}H HSQC spectra of LC8 prepared with different Cu(II)-EDTA dopant concentration

	Δδ (ppm) of 0mM vs. 5mM Cu(II)-EDTA		Δδ (ppm) of 0mM vs. 10mM Cu(II)-EDTA		Δδ (ppm) of 0mM vs. 20mM Cu(II)-EDTA		Δδ (ppm) of 0mM vs. 50mM Cu(II)_EDTA	
Residue	N	H	N	H	N	H	N	H
M1	0.027	0.005	0.019	0.007	0.047	0.011	0.078	0.015
S2	0.012	0.005	-0.007	0.005	-0.003	0.007	0.009	0.008
D3	0.003	0.006	-0.022	0.008	-0.020	0.013	-0.021	0.017
R4	0.037	0.004	0.027	0.007	0.054	0.010	0.073	0.011
K5	0.030	0.008	0.035	0.016	0.082	0.028	0.132	0.038
A6	0.005	0.004	-0.025	0.004	-0.029	0.006	-0.025	0.009
V7	0.034	0.007	0.017	0.009	0.051	0.013	0.079	0.018
I8	0.000	0.001	-0.028	-0.001	-0.027	-0.001	-0.030	-0.004
K9	0.047	0.002	0.088	0.008	0.121	0.011	0.150	0.012
N10	-0.008	0.001	-0.053	0.004	-0.081	0.007	-0.100	0.007
A11	0.000	0.000	0.003	-0.004	-0.001	0.002	0.008	0.017
D12	**-0.011**	**0.002**	**-0.009**	**-0.001**	**ND***	**ND**	**ND**	**ND**
M13	**-0.106**	**-0.013**	**0.146**	**-0.015**	**ND**	**ND**	**ND**	**ND**
S14	**-0.080**	**-0.002**	**0.143**	**-0.048**	**ND**	**ND**	**ND**	**ND**
E15	-0.023	-0.005	-0.052	-0.006	-0.076	-0.011	-0.092	-0.015
E16	-0.012	-0.010	-0.033	-0.013	-0.042	-0.026	-0.051	-0.035
M17	-0.002	-0.007	-0.025	-0.003	-0.023	-0.005	-0.020	-0.003
Q18	-0.025	0.003	-0.049	0.003	-0.075	0.002	-0.078	0.004
Q19	0.031	0.004	-0.003	0.006	-0.014	0.008	-0.032	0.010
D20	0.000	0.006	-0.009	0.015	-0.003	0.025	-0.023	-0.002
A21	0.020	-0.002	0.017	-0.002	0.057	-0.001	0.068	0.000
V22	-0.043	-0.005	-0.057	-0.005	-0.071	-0.008	-0.073	-0.011
D23	0.009	0.004	-0.014	0.009	-0.009	0.014	-0.022	0.017
C24	-0.025	0.001	-0.052	0.001	-0.063	0.002	-0.077	0.003
A25	-0.010	-0.002	-0.015	-0.003	-0.013	-0.006	-0.002	-0.009
T26	-0.024	0.004	-0.045	0.006	-0.058	0.010	-0.054	0.012
Q27	0.015	0.004	-0.027	0.010	-0.007	0.015	-0.008	0.020
A28	0.024	-0.002	-0.015	-0.002	-0.017	-0.001	-0.017	-0.003
L29	0.016	0.006	0.015	0.011	0.022	0.017	0.045	0.021
E30	-0.003	0.000	-0.032	0.005	-0.039	0.008	-0.030	0.012
K31	0.018	0.004	-0.001	0.007	0.002	0.010	0.023	0.013
Y32	0.008	-0.001	-0.007	0.001	-0.003	0.001	0.002	0.000
N33	0.061	0.011	0.038	0.016	0.064	0.027	0.092	0.034
I34	0.038	0.009	0.054	0.014	0.101	0.024	0.133	0.030
E35	**-0.054**	**-0.015**	**-0.113**	**-0.025**	**-0.145**	**-0.050**	**-0.223**	**-0.072**
K36	**0.103**	**0.007**	**0.190**	**0.015**	**0.307**	**0.026**	**0.487**	**0.029**
D37	-0.097	-0.009	-0.261	-0.014	-0.031	-0.022	-0.011	0.010
I38	**-0.057**	**-0.004**	**-0.118**	**0.004**	**-0.188**	**0.003**	**-0.237**	**0.004**
A39	**0.084**	**0.009**	**0.122**	**0.012**	**0.209**	**0.023**	**0.275**	**0.026**
A40	0.003	-0.022	-0.021	-0.017	-0.018	-0.012	-0.031	-0.009
Y41	-0.037	0.003	-0.056	0.007	-0.083	0.010	-0.080	0.016
I42	0.035	0.002	0.014	0.009	0.018	0.013	0.070	0.020
K43	0.012	0.007	0.031	0.015	0.055	0.027	0.095	0.033
K44	-0.050	-0.004	-0.115	-0.005	-0.154	-0.005	-0.208	-0.002
E45	0.002	0.005	-0.031	0.007	-0.052	0.013	-0.056	0.022
F46	0.004	0.004	0.000	0.010	0.032	0.015	0.066	0.019

D47	-0.003	-0.008	-0.058	-0.011	-0.050	-0.016	-0.107	-0.024
K48	-0.024	-0.006	-0.059	-0.003	-0.114	-0.008	-0.168	-0.009
K49	0.019	0.004	0.030	0.009	0.066	0.015	0.100	0.022
Y50	-0.042	0.001	-0.011	0.001	-0.025	0.008	-0.135	0.002
N51	0.029	-0.009	0.022	-0.005	0.083	-0.004	0.048	0.004
T53	**0.026**	**-0.015**	**0.005**	**-0.015**	**0.083**	**-0.037**	**0.222**	**-0.026**
W54	**0.046**	**-0.004**	**0.068**	**-0.005**	**0.140**	**-0.009**	**0.228**	**-0.004**
H55	-0.011	-0.004	-0.043	-0.008	-0.052	-0.020	-0.053	-0.029
C56	0.000	0.004	-0.006	0.014	-0.028	0.031	-0.062	0.038
I57	**-0.051**	**0.008**	**-0.127**	**0.007**	**-0.209**	**0.012**	**-0.279**	**0.011**
V58	-0.031	0.010	-0.080	0.031	-0.101	0.057	-0.136	0.078
G59	-0.016	0.009	-0.060	0.018	-0.078	0.026	-0.127	0.038
R60	0.001	0.003	-0.057	0.009	-0.045	0.015	-0.070	0.020
N61	0.016	0.015	-0.003	0.024	0.008	0.039	0.029	0.054
F62	0.051	0.007	0.063	0.019	0.111	0.032	0.145	0.040
G63	0.050	0.017	0.060	0.027	0.066	0.050	0.114	0.066
S64	**0.071**	**0.009**	**0.099**	**0.000**	**ND**	**ND**	**ND**	**ND**
Y65	-0.180	-0.040	0.302	0.040	-0.187	-0.033	-0.190	-0.031
V66	0.094	-0.025	0.061	0.010	0.111	-0.018	-0.008	-0.016
T67	**0.003**	**-0.013**	**-0.038**	**-0.007**	**ND**	**ND**	**ND**	**ND**
E69	**0.093**	**0.005**	**0.000**	**0.003**	**ND**	**ND**	**ND**	**ND**
T70	**0.102**	**0.004**	**-0.814**	**0.002**	**-0.777**	**0.011**	**-0.751**	**0.017**
R71	**0.093**	**0.006**	**-0.200**	**-0.018**	**ND**	**ND**	**ND**	**ND**
H72	**-0.058**	**0.015**	**-0.075**	**-0.004**	**0.082**	**-0.017**	**ND**	**ND**
F73	-0.108	-0.006	0.124	0.002	0.150	0.002	0.073	0.014
I74	-0.007	-0.004	-0.031	-0.007	0.056	0.007	0.040	0.017
Y75	0.019	0.006	-0.017	0.011	-0.009	0.016	0.013	0.031
F76	-0.010	0.006	-0.025	0.010	-0.038	0.016	-0.043	0.020
Y77	-0.037	0.006	-0.076	0.006	-0.120	0.012	-0.146	0.018
L78	-0.012	0.010	-0.023	0.011	-0.019	0.017	-0.022	0.019
G79	0.001	0.004	-0.029	0.008	-0.033	0.013	-0.029	0.018
Q80	-0.025	0.010	-0.033	0.008	-0.043	0.010	-0.026	0.011
V81	0.008	0.005	-0.016	0.010	-0.013	0.015	-0.026	0.018
A82	0.023	0.004	0.016	0.014	0.039	0.021	0.049	0.028
I83	0.019	0.010	0.000	0.013	0.016	0.019	0.023	0.023
L84	0.009	0.005	-0.022	0.009	-0.007	0.015	-0.015	0.017
L85	-0.001	-0.003	-0.022	-0.008	-0.021	-0.015	-0.028	-0.023
F86	-0.017	0.002	-0.041	-0.001	-0.054	-0.002	-0.069	-0.017
K87	-0.083	-0.005	NA	NA	NA	NA	NA	NA

*ND- not detectable because of paramagnetically-induced broadening

Table S2. Solid-state chemical shift perturbations observed in the NCACB spectra of LC8 samples containing different Cu(II)-EDTA dopant concentrations. Solid-state chemical shifts of the LC8 sample doped with 5 mM Cu(II)-EDTA are shown. The data presented in this table are also plotted as bar charts in Figure S3, where it can be clearly seen that the extent of chemical shift perturbations and the number of peaks whose shifts are perturbed increase as a function of the paramagnetic dopant concentration.

	Δδ, 0mM vs. 5mM Cu(II)-EDTA			Δδ, 0mM vs. 10mM Cu(II)-EDTA			Δδ, 0mM vs. 50mM Cu(II)-EDTA			δ, 5 mM Cu(II)-EDTA		
Residue	N	C^{α}	C^{β}	N	C^{α}	C^{β}	N	C^{α}	C^{β}	N	C^{α}	C^{β}
M1												
S2												
D3												
R4												
K5												
A6												
V7	-0.057	0.007	-0.052	-0.219	-0.010	-0.067	-0.208	0.015	-0.178	123.271	61.85	34.004
I8	-0.084	-0.014	-0.037	-0.256	0.080	0.224	-0.134	-0.372	-0.184	128.893	61.222	36.212
K9	0.063	0.054	0.002	-0.059	0.152	-0.009	0.337	0.194	0.110	129.427	56.173	32.418
N10	-0.050	0.096	0.011	-0.215	0.146	0.522	NA	NA	NA	114.8385	53.9195	42.858
A11	0.356	-0.046	0.011	0.335	-0.026	-0.018	0.663	-0.151	-0.073	124.104	51.937	22.471
D12	0.059	-0.079	-0.039	0.086	-0.120	0.116	NA	NA	NA	122.293	53.139	41.226
M13	-0.234	0.140	0.159	-0.804	0.104	0.205	NA	NA	NA	120.928	56.549	39.107
S14	-0.585	-0.101	0.039	-1.262	-0.199	-0.209	NA	NA	NA	124.361	58.382	64.169
E15	-0.020	0.044	0.020	0.152	0.165	-0.010	-0.084	0.134	-0.204	122.954	60.442	29.204
E16												
M17	-0.229	-0.084	0.132	-0.297	-0.026	0.027	-0.544	0.275	-0.374	120.895	59.661	33.042
Q18	0.009	-0.059	0.028	0.304	0.026	-0.012	1.247	-0.729	0.233	118.728	59.616	29.079
Q19	0.069	0.031	-0.005	0.130	0.135	0.012	-0.220	0.026	0.032	116.683	57.896	28.004
D20												
A21	0.020	-0.078	0.005	-0.293	0.022	-0.139	-0.189	0.047	-0.054	124.96	55.9325	17.62
V22	-0.083	0.011	0.002	-0.290	-0.072	-0.052	-0.426	-0.030	-0.045	118.88	66.831	31.814
D23	-0.171	-0.003	0.036	-0.012	0.013	0.045	-0.055	-0.029	0.097	123.833	57.803	40.193
C24	-0.104	-0.010	-0.040	-0.214	0.033	-0.084	-0.334	0.132	-0.058	121.163	62.927	27.61

A25	-0.045	0.011	-0.009	0.147	0.064	-0.092	0.296	0.151	-0.100	120.238	55.017	19.963
T26	-0.142	-0.014	-0.038	-0.075	0.032	-0.042	-0.028	0.008	0.007	115.274	67.804	68.729
Q27	0.020	-0.031	0.017	-0.030	-0.018	-0.028	-0.168	-0.074	-0.120	120.278	58.625	28.529
A28	-0.112	0.007	-0.043	-0.117	0.006	-0.084	-0.006	0.099	-0.129	122.473	55.178	20.961
L29	-0.007	0.018	0.025	-0.081	0.089	-0.062	0.197	0.259	0.125	116.659	56.991	42.069
E30	0.045	0.001	-0.005	-0.002	0.009	-0.133	-0.337	0.061	-0.031	117.252	58.39	29.614
K31	-0.086	0.020	0.018	0.092	-0.091	0.030	0.143	-0.027	-0.019	117.622	58.174	35.498
Y32												
N33	0.016	0.062	0.064	0.465	0.075	0.019	-0.400	-0.064	-0.212	110.208	53.32	41.852
I34	-0.089	0.137	0.027	0.455	-0.004	0.401	NA	NA	NA	122.009	60.665	39.375
E35	-0.304	-0.058	0.034	NA	NA	NA	NA	NA	NA	129.729	62.523	28.319
K36	0.309	0.014	0.081	0.948	-0.084	0.510	NA	NA	NA	116.167	59.764	31.743
D37												
I38	-0.205	0.016	-0.040	-0.353	0.033	-0.065	-0.139	0.081	-0.090	122.905	65.485	38.403
A39	0.029	0.026	0.048	0.217	0.074	0.121	0.464	0.177	-0.032	119.016	54.811	17.891
A40	0.029	0.026	0.048	0.217	0.074	0.121	0.464	0.177	-0.032	119.016	54.811	17.891
Y41	-0.026	0.049	0.034	0.198	0.090	0.011	-0.026	0.107	-0.035	117.008	61.55	38.448
I42	-0.131	0.046	0.018	-0.142	0.024	-0.029	-0.101	0.108	-0.029	117.812	64.871	38.386
K43	0.058	0.009	0.078	-0.293	-0.020	-0.373	-0.006	-0.155	-0.033	118.647	61.127	35.876
K44												
E45	-0.205	0.066	0.072	-0.097	-0.010	-0.048	0.025	0.051	-0.246	119.736	58.961	29.602
F46	-0.104	-0.029	-0.041	-0.259	-0.121	-0.099	0.273	-0.120	-0.255	122.891	63.737	35.658
D47												
K48	-0.162	0.008	0.026	0.007	-0.041	0.076	-0.457	0.393	-0.274	117.443	59.037	32.818
K49	-0.308	-0.041	0.038	-0.145	-0.084	0.025	-0.427	-0.073	-0.293	117.513	58.828	34.055
Y50	0.010	-0.041	-0.055	0.221	0.242	0.097	0.149	0.326	0.320	113.566	58.182	40.283
N51	-0.266	-0.010	0.005	-0.129	-0.142	0.146	0.425	-0.053	0.015	113.628	55.372	36.381
P52	0.048	-0.119	-0.104	0.342	-0.094	-0.096	0.810	-0.471	-0.225	128.819	60.252	32.272
T53	-0.082	-0.078	0.047	-0.031	-0.130	0.145	0.402	0.012	0.136	122.224	64.861	69.986
W54	0.067	0.000	-0.002	0.229	-0.004	-0.094	0.454	-0.039	-0.185	128.425	55.85	30.145
H55	-0.084	-0.017	0.096	0.025	-0.132	0.155	-0.548	-0.363	-0.133	118.3	54.863	34.353
C56	0.024	-0.024	0.047	0.056	-0.013	-0.063	-0.229	-0.120	0.147	120.354	56.588	31.097
I57	-0.272	-0.131	-0.070	-0.278	0.005	-0.077	-0.721	-0.333	-0.254	132.1785	59.794	39.824

V58	-0.014	0.043	-0.048	-0.080	0.043	-0.144	-0.350	0.060	-0.072	124.377	59.415	36.744
G59	0.047	-0.045		0.112	0.091		-0.483	0.037		112.257	46.755	
R60												
N61												
F62	0.019	0.022	-0.099	0.228	0.016	-0.079	0.157	-0.048	-0.220	119.076	57.173	40.15
G63	-0.013	0.059		-0.132	0.104		0.252	0.254		107.931	43.575	
S64	0.078	-0.001	-0.017	0.466	-0.142	-0.191	NA	NA	NA	111.256	57.249	68.509
Y65										122.368		
V66	0.078	0.034	-0.083	-0.214	-0.191	-0.255	NA	NA	NA	119.933	57.389	33.311
T67	-0.220	0.108	0.100	0.498	0.098	-0.079	NA	NA	NA	122.332	61.608	70.056
H68												
E69												
T70	0.220	-0.013	0.011	0.915	-0.255	0.050	NA	NA	NA	125.834	64.175	68.879
R71												
H72												
F73	0.249	-0.183	0.151	0.606	-0.243	0.358	NA	NA	NA	120.361	58.124	43.302
I74	-0.190	0.041	-0.003	-0.108	-0.112	-0.055	-0.056	0.353	-0.209	128.615	61.814	40.616
Y75	0.058	0.084	0.042	0.208	0.077	0.202	0.198	-0.024	0.342	126.973	54.388	40.906
F76	-0.073	-0.042	-0.106	-0.139	0.027	-0.060	0.321	0.218	-0.312	125.016	54.77	43.091
Y77	0.054	-0.307	-0.036	-0.155	-0.433	-0.189	-0.804	-0.138	0.302	118.052	55.727	41.851
L78												
G79	-0.365	-0.004		-0.525	-0.043		-0.567	-0.007		114.317	46.777	
Q80	-0.030	-0.070	-0.058	0.042	0.093	-0.013	0.092	0.271	0.292	125.044	56.05	29.505
V81	-0.072	-0.054	0.078	-0.356	0.033	0.002	-0.087	0.193	-0.145	119.462	62.667	33.165
A82	0.033	0.028	-0.002	0.235	0.138	-0.002	0.459	0.128	-0.087	128.695	50.262	20.656
I83	0.079	-0.024	-0.037	0.104	0.074	-0.101	-0.101	0.124	-0.175	119.631	60.551	39.763
L84	0.137	0.093	0.045	0.230	-0.014	-0.075	0.703	0.200	0.092	127.722	53.164	46.368
L85	-0.229	0.133	-0.026	-0.102	0.028	-0.080	-0.051	-0.349	-0.195	129.913	53.957	46.576
F86	-0.169	-0.128	-0.015	0.060	0.047	0.064	NA	NA	NA	120.946	56.166	42.156
K87												
S88												
G89												

Table S3 Chemical shift differences observed in a pair of 3D NCACB spectra (NUS and US) for LC8 doped with 5 mM Cu(II)-EDTA. The US spectrum was acquired as a 32 x 32 point hypercomplex matrix in indirect dimensions, and in the NUS data 25% (256) hypercomplex points of the US schedule were sampled. The digital resolution is the same for both spectra.

	$\Delta\delta$ (C^{α})	$\Delta\delta$ (N)	$\Delta\delta$ (C^{β})
M1			
S2			
D3			
R4			
K5			
A6			
V7	0.160	0.164	0.129
I8	0.028	0.053	-0.006
K9	0.035	0.131	0.029
N10	0.077	0.060	0.112
A11	0.028	0.127	0.064
D12	-0.076	0.028	-0.013
M13	0.063	0.034	-0.095
S14	-0.005	-0.262	0.061
E15	0.000	0.085	0.008
E16			
M17	0.017	0.031	0.070
Q18	0.058	-0.129	-0.021
Q19	-0.027	0.030	0.075
D20			
A21	-0.033	0.231	-0.052
V22	0.028	0.016	0.032
D23	-0.049	-0.138	-0.025
C24	0.036	0.039	0.010
A25	-0.005	0.015	0.012
T26	0.034	0.048	-0.005
Q27	-0.009	-0.014	0.006
A28	0.030	0.062	0.016
L29	0.020	-0.030	0.017
E30	-0.011	0.012	0.018
K31	-0.141	0.110	-0.041
Y32			
N33	0.104	-0.055	0.057
I34	0.121	0.134	0.029
E35	-0.012	-0.139	-0.034
K36	0.016	0.158	0.100
D37			
I38	0.037	-0.068	-0.018
A39	0.037	0.051	0.048
A40	0.037	0.051	0.048
Y41	0.004	-0.014	0.025
I42	0.051	0.013	0.025

K43	0.008	0.093	0.026
K44			
E45	0.058	-0.009	0.101
F46	-0.051	-0.016	-0.029
D47			
K48	0.143	0.021	-0.074
K49	0.043	0.101	0.102
Y50	-0.041	0.064	0.242
N51	0.015	0.051	-0.052
P52	0.003	0.092	-0.061
T53	0.110	0.022	-0.011
W54	0.007	0.070	-0.017
H55	0.026	-0.011	0.096
C56	0.016	0.025	0.047
I57	0.014	-0.006	0.014
V58	0.001	0.056	0.063
G59	0.006	-0.002	-0.021
R60			
N61			
F62	0.010	0.092	-0.005
G63	0.016	0.055	0.040
S64	0.085	0.015	-0.005
Y65			
V66	0.073	0.067	-0.031
T67	0.111	-0.149	0.137
H68			
E69			
T70	0.046	-0.176	0.028
R71			
H72			
F73	-0.127	0.035	0.113
I74	0.049	-0.003	0.063
Y75	0.049	0.108	0.002
F76	-0.078	0.022	0.125
Y77	0.143	-0.025	0.167
L78			
G79	0.047	0.067	0.008
Q80	-0.071	0.013	-0.095
V81	-0.018	0.077	0.071
A82	-0.040	-0.087	0.005
I83	0.007	0.011	0.061
L84	0.106	0.121	0.104
L85	0.057	-0.071	0.091
F86	-0.040	0.017	0.021
K87			
S88			
G89			

Table S4. The Chemical Shifts Assignments of LC8 on the Basis of NUS-PACC Experiments Conducted on the Sample Doped with 5 mM Cu(II)-EDTA.*

Residue	N	C^{α}	C^{β}	C'
M1				
S2				
D3				
R4				
K5				
A6		51.4		177.3
V7	123.1	61.7	34.0	175.3
I8	128.8	61.3	36.2	176.1
K9	129.3	56.1	32.4	176.8
N10	114.8	53.9	42.9	172.1
A11	124.0	52.0	22.5	175.2
D12	122.4	53.2	41.2	173.6
M13	120.9	56.2	39.1	175.0
S14	123.9	58.4	64.2	174.6
E15	123.0	60.5	29.2	179.1
E16				
M17	120.8	59.7	33.0	177.7
Q18	118.7	59.5	29.1	177.8
Q19	116.7	57.9	28.0	
D20				
A21	125.0	56.0	17.6	178.6
V22	118.9	66.8	31.8	179.8
D23	123.8	57.8	40.2	178.5
C24	121.1	62.9	27.6	176.7
A25	120.2	55.0	20.0	178.1
T26	115.3	67.8	68.7	176.8
Q27	120.2	58.7	28.5	178.7
A28	122.6	55.1	21.0	178.5
L29	116.8	57.0	42.1	179.2
E30	117.2	58.4	29.6	178.1
K31	117.5	58.2	35.5	
Y32		56.5		173.5
N33	110.3	53.3	41.9	176.0
I34	122.0	61.0	39.4	177.9
E35	129.7	62.4	28.3	178.2
K36	116.1	59.8	31.7	177.2
D37		56.4		178.5
I38	123.0	65.5	38.4	176.7
A39	119.0	54.8	17.9	178.5
A40	119.1	54.9	17.9	178.4
Y41	117.0	61.6	38.4	177.6
I42	117.8	64.9	38.4	177.1
K43	118.6	61.0	35.9	178.7
K44	116.9	60.0		180.4
E45	119.8	58.8	29.6	178.7

F46	123.0	63.7	35.7	177.8
D47				
K48	117.4	58.9	32.8	178.0
K49	117.6	58.9	34.1	177.8
Y50	113.8	58.2	40.3	176.8
N51	113.7	55.4	36.4	172.4
P52	128.4	60.0	32.3	178.4
T53	122.3	64.8	70.0	172.1
W54	128.3	55.9	30.1	174.0
H55	118.3	54.9	34.4	174.7
C56	120.4	56.6	31.1	171.2
I57	132.2	59.8	39.8	173.5
V58	124.4	59.4	36.7	175.5
G59	112.3	46.8		172.6
R60				
N61				
F62	119.1	57.5	40.2	173.4
G63	107.8	43.6		171.6
S64	111.2	57.2	68.5	171.9
Y65	122.1	61.4		173.2
V66	119.9	57.1	33.3	175.0
T67	122.4	61.6	70.1	175.0
H68				
E69				
T70	125.9	64.0	68.9	
R71				
H72				
F73	120.4	58.2	43.3	172.4
I74	128.6	61.8	40.6	169.0
Y75	126.9	54.5	40.9	174.4
F76	125.0	54.8	43.1	169.8
Y77	118.0	55.7	41.9	176.4
L78				
G79	114.5	46.8		175.3
Q80	125.1	56.1	29.5	174.8
V81	119.4	62.6	33.2	172.1
A82	128.7	50.3	20.7	176.0
I83	119.7	60.6	39.8	173.2
L84	127.7	53.1	46.4	173.8
L85	130.0	53.9	46.6	174.1
F86	120.9	56.3	42.2	172.9
K87	116.4	54.9	34.4	
S88				
G89				

*The chemical shifts for N C^{α}, and C' are the average values of chemical shifts extracted from 3D NUS-PACC NCACB, NCACO and NCOCA spectra.

Table S5. The standard deviations for the resolved residues in 3D NUS-PACC NCOCA spectra acquired three times. The three NCOCA spectra were collected back to back with the same settings and the sampling schedules shown in Figure 2 (B). Spectra were processed by MINT; 90 degree sinebell apodization was used for the direct dimension.

	C' (ppm)	N (ppm)	CA (ppm)
M1			
S2			
D3			
R4			
K5			
A6	0.11		0.09
V7	0.03	0.04	0.02
I8	0.02	0.05	0.02
K9	0.05	0.07	0.02
N10	0.05	0.07	0.03
A11	0.03	0.08	0.03
D12		0.19	
M13	0.01		0.01
S14	0.04	0.01	0.02
E15		0.03	
E16			
M17	0.04		0.04
Q18	0.02	0.01	0.06
Q19		0.04	
D20			
A21	0.03		0.03
V22	0.06	0.02	0.01
D23	0.01	0.07	0.03
C24	0.03	0.01	0.02
A25	0.05	0.02	0.01
T26	0.02	0.06	0.02
Q27	0.03	0.04	0.04
A28	0.04	0.05	0.03
L29	0.08	0.04	0.02
E30	0.06	0.01	0.05
K31		0.02	
Y32	0.06		0.04
N33		0.08	
I34	0.04		0.10
E35	0.10	0.02	0.04
K36		0.04	
D37	0.02		0.01
I38	0.03	0.06	0.02
A39	0.05	0.05	0.01
A40	0.04	0.04	0.03
Y41	0.04	0.04	0.01
I42	0.02	0.04	0.03
K43		0.05	

K44	0.04		0.07
E45	0.03	0.06	0.04
F46		0.05	
D47			
K48			
K49	0.01		0.03
Y50	0.07	0.04	0.04
N51	0.07	0.08	0.07
P52	0.00	0.03	0.02
T53	0.02	0.06	0.04
W54	0.02	0.03	0.03
H55	0.01	0.05	0.01
C56	0.05	0.01	0.01
I57	0.03	0.01	0.02
V58	0.04	0.04	0.03
G59		0.01	
R60			
N61			
F62	0.04		0.07
G63	0.06	0.05	0.06
S64	0.03	0.16	0.02
Y65	0.04	0.11	0.22
V66	0.05	0.07	0.04
T67		0.14	
H68			
E69			
T70			
R71			
H72			
F73	0.06		0.03
I74	0.02	0.08	0.03
Y75	0.01	0.04	0.03
F76	0.01	0.05	0.02
Y77		0.05	
L78			
G79	0.11		0.03
Q80	0.05	0.04	0.03
V81	0.00	0.06	0.06
A82	0.04	0.05	0.02
I83	0.04	0.02	0.03
L84	0.03	0.01	0.05
L85	0.02	0.07	0.01
F86		0.01	
K87			
S88			
G89			

Table S6. The NUS Schedules for the Three 3D Experiments

	NCOCA		NCACB		NCACO	
	t_1	t_2	t_1	t_2	t_1	t_2
1	1	1	1	1	1	1
2	2	1	2	3	1	2
3	2	4	6	1	1	7
4	3	3	2	1	3	2
5	4	2	2	7	2	6
6	9	1	1	3	1	9
7	7	1	3	5	4	1
8	2	5	10	4	1	8
9	1	8	5	6	7	4
10	6	7	10	1	1	4
11	9	4	4	5	5	10
12	9	2	6	3	8	6
13	5	6	5	1	7	1
14	1	6	3	11	7	9
15	7	4	8	1	11	5
16	10	6	11	7	6	5
17	5	8	11	4	3	8
18	4	4	15	3	8	1
19	1	10	1	15	12	3
20	14	2	7	5	15	2
21	1	3	6	10	9	4
22	1	11	9	8	7	5
23	2	15	4	8	5	3
24	2	9	12	8	13	5
25	1	13	1	11	14	2
26	7	5	10	7	9	7
27	11	1	13	7	12	7
28	8	1	1	9	6	9
29	3	12	10	3	7	14
30	3	7	13	3	7	12
31	8	6	3	9	1	19
32	7	6	12	4	4	5
33	12	4	7	6	2	3
34	2	7	5	12	6	13
35	1	9	12	7	8	9
36	4	6	7	12	16	3
37	4	12	2	17	3	11
38	8	7	12	10	2	9
39	9	3	1	12	5	4
40	14	6	16	1	12	1
41	2	17	15	6	7	3
42	7	10	3	18	3	20
43	6	9	12	1	6	7
44	3	14	9	3	10	13
45	4	8	4	4	11	4

46	4	1	14	6	14	3
47	2	14	5	13	6	18
48	11	5	4	13	10	14
49	13	6	18	3	3	23
50	16	6	18	8	8	8
51	3	8	7	14	8	7
52	21	1	19	7	4	2
53	5	9	20	4	8	15
54	14	7	12	11	12	12
55	22	1	2	4	13	8
56	14	4	4	2	3	10
57	8	2	2	18	19	3
58	6	16	25	3	20	3
59	12	2	2	12	5	8
60	12	3	5	4	12	9
61	16	5	21	3	8	12
62	7	9	3	20	5	6
63	18	1	2	16	8	16
64	7	14	24	1	11	6
65	22	3	24	2	13	11
66	4	17	10	17	21	2
67	10	4	10	5	8	5
68	3	11	3	17	17	4
69	9	11	8	2	5	5
70	3	6	12	16	4	22
71	20	5	13	1	8	3
72	4	21	25	4	12	4
73	2	16	17	11	14	4
74	13	8	17	3	5	1
75	20	7	7	3	1	12
76	5	19	25	2	19	1
77	20	8	10	10	10	10
78	9	12	8	5	2	1
79	12	14	11	8	21	3
80	13	9	5	17	12	5
81	6	18	3	25	4	24
82	15	6	6	5	21	4
83	13	2	14	16	10	18
84	5	21	15	14	18	3
85	6	13	3	7	11	11
86	20	2	4	20	2	8
87	19	5	9	9	7	2
88	4	3	14	4	4	14
89	8	10	2	13	5	24
90	29	1	2	23	10	9
91	15	4	29	2	9	11
92	4	11	16	11	8	17
93	14	9	7	7	11	17
94	10	13	6	9	2	18

95	6	12	22	7	16	12
96	9	9	1	21	2	23
97	19	4	6	21	1	22
98	17	9	14	3	8	13
99	5	12	17	12	15	14
100	15	8	1	25	17	2
101	14	8	20	1	4	10
102	7	17	11	5	7	7
103	21	9	1	14	24	4
104	1	5	9	17	14	10
105	13	5	6	13	3	18
106	3	1	15	12	7	21
107	3	10	18	15	1	6
108	24	5	5	19	5	20
109	17	4	14	1	3	16
110	3	17	21	12	21	5
111	1	4	10	14	4	20
112	14	15	15	11	23	3
113	7	16	20	10	10	1
114	18	7	1	19	15	13
115	1	12	9	7	16	4
116	10	1	19	15	14	16
117	12	17	19	11	20	1
118	15	1	32	1	5	21
119	4	20	23	4	17	14
120	7	13	6	26	14	1
121	31	1	12	6	18	1
122	25	5	19	8	19	10
123	27	3	4	15	21	6
124	4	18	15	5	9	1
125	17	3	24	10	2	4
126	4	9	17	5	5	17
127	22	4	15	8	6	11
128	23	4	15	18	12	11
129	3	27	1	31	24	5
130	5	17	2	9	18	12
131	6	1	12	15	11	12
132	27	6	8	3	9	23
133	7	21	16	6	16	11
134	22	7	16	8	13	20
135	16	10	8	11	19	12
136	9	6	2	22	7	22
137	4	26	21	14	10	11
138	12	9	19	9	14	9
139	10	9	30	6	8	4
140	9	7	7	9	20	6
141	1	14	2	2	25	5
142	13	4	2	31	18	10
143	13	11	20	13	3	13

144	27	7	4	26	10	7
145	12	7	15	4	3	19
146	24	1	25	9	10	19
147	12	15	9	12	13	22
148	9	20	29	4	6	8
149	17	12	23	5	12	24
150	9	14	1	8	1	21
151	8	24	30	1	18	8
152	32	2	16	13	6	23
153	4	28	7	10	26	5
154	14	18	9	1	16	18
155	7	25	8	4	22	4
156	2	18	2	30	16	13
157	13	13	8	25	5	12
158	16	9	15	21	15	11
159	28	6	18	5	24	7
160	13	14	22	4	19	9
161	5	5	17	8	14	21
162	28	5	2	21	1	3
163	14	13	23	15	9	3
164	3	21	7	13	13	18
165	4	25	8	10	26	1
166	7	18	2	10	8	24
167	27	5	20	2	7	16
168	19	10	9	23	17	9
169	31	2	17	14	6	10
170	28	1	25	1	9	15
171	15	5	8	22	5	15
172	14	1	20	14	30	2
173	9	18	4	25	10	16
174	26	6	26	10	13	24
175	3	16	19	5	6	3
176	1	20	10	18	28	1
177	2	23	10	6	12	16
178	23	6	10	12	17	11
179	8	18	1	18	8	14
180	8	14	30	9	11	21
181	6	5	23	3	18	11
182	22	9	7	26	9	22
183	12	22	24	6	12	18
184	22	12	18	20	2	21
185	23	1	4	27	13	19
186	9	23	5	16	4	8
187	3	29	21	4	3	6
188	15	15	9	16	13	10
189	14	11	4	16	31	2
190	17	10	5	28	10	22
191	31	4	10	29	23	4
192	1	30	15	20	32	24

193	20	11	18	2
194	20	15	14	2
195	17	14	11	10
196	16	7	15	16
197	14	14	6	7
198	21	10	3	30
199	1	19	9	30
200	30	2	14	9
201	19	2	24	18
202	4	31	8	6
203	8	15	30	2
204	8	4	5	18
205	3	32	9	4
206	8	13	14	22
207	20	4	23	6
208	8	11	20	6
209	26	7	27	11
210	12	19	14	8
211	5	14	12	24
212	16	18	3	14
213	19	9	7	27
214	17	13	24	11
215	1	7	19	6
216	1	29	9	27
217	13	24	7	2
218	20	17	27	13
219	27	9	12	17
220	10	2	17	9
221	16	12	4	12
222	17	8	25	17
223	6	3	30	7
224	8	26	16	14
225	19	16	16	27
226	17	7	3	24
227	7	7	7	11
228	30	6	11	20
229	4	32	12	29
230	32	4	17	6
231	11	7	1	27
232	2	22	21	15
233	5	11	24	14
234	10	25	2	28
235	27	8	3	32
236	5	28	1	20
237	16	4	17	1
238	21	3	17	13
239	13	22	5	8
240	31	7	18	7
241	20	1	20	16

242	3	9	29	9
243	11	14	20	12
244	2	10	14	15
245	23	3	26	9
246	4	30	6	8
247	18	3	23	16
248	14	24	9	13
249	2	2	13	4
250	27	4	5	31
251	10	15	8	15
252	18	21	11	19
253	5	26	29	8
254	23	12	5	20
255	15	16	23	18
256	32	32	32	32