

**Table 3. Extracted values of  $D_{\text{NH}}$ ,  $D_{\text{NH}}^{\text{A}}$ ,  $D_{\text{NH}}^{\text{B}}$ ,  $\sigma_{\text{N}}$  and reduced  $\chi^2$  obtained from fits of relaxation dispersion data recorded on G48M Fyn SH3, ~ 35 mg/ml Pf1 phage, 25°C.**

Residue	$D_{\text{NH}}(\text{Hz})$	err	$D_{\text{NH}}^{\text{A}}$	err	$D_{\text{NH}}^{\text{B}}$	err	(ppm)	err	$\chi^2_{\text{red}}$	$R_{\text{zz}}-R_1$
5	1.6	5.2	4.9	0.3	3.3	5.2	-3.54	0.05	1.1	1.1
7	3.6	10.0	-3.6	0.3	-7.2	10.0	5.95	0.05	1.3	1.3
8	-28.9	11.0	-12.3	0.3	16.6	11.0	-8.01	0.05	1.4	1.4
9	-15.4	4.9	-17.2	0.3	-1.7	4.9	-4.53	0.05	1.1	0.9
10	-26.4	2.8	-20.9	0.3	5.5	2.8	2.57	0.05	1.2	0.7
11	-14.0	7.8	-15.0	0.3	-1.0	7.8	5.77	0.05	1.5	1.6
12	-11.0	1.8	-11.8	0.3	-0.8	1.8	2.01	0.05	0.7	1.1
13	4.5	2.1	4.5	0.3	-0.1	2.1	1.76	0.05	1.0	5.3
14	13.2	6.5	4.0	0.3	-9.2	6.5	-4.16	0.05	1.7	6.8
15	3.0	12.0	4.1	0.3	1.1	12.0	-3.32	0.05	1.8	22.6
17	23.1	11.2	-0.5	0.3	-23.5	11.2	-0.06	0.05	0.5	2.0
19	-6.1	1.5	-4.3	0.3	1.8	1.5	-0.63	0.05	0.5	1.2
20	-22.7	9.4	-22.7	0.3	0.1	9.4	-6.43	0.05	1.5	1.4
21	-17.0	2.0	-21.2	0.3	-4.2	2.0	-2.16	0.05	1.1	2.3
22	-1.5	3.1	-1.5	0.3	-0.0	3.1	0.26	0.05	0.5	1.9
23	-8.2	5.7	-3.5	0.3	4.7	5.7	6.07	0.05	1.0	2.1
24	-7.9	2.8	-8.7	0.3	-0.7	2.8	3.39	0.05	0.9	1.4
26	14.0	3.2	12.1	0.3	-1.9	3.2	-3.44	0.05	0.9	1.4
27	26.9	1.6	24.2	0.3	-2.7	1.7	-0.73	0.05	0.6	1.1
28	33.3	6.7	22.6	0.3	-10.7	6.7	4.10	0.05	1.8	0.8
29	14.3	1.7	7.8	0.3	-6.5	1.7	1.92	0.05	1.0	1.6
30	12.8	4.6	0.1	0.3	-12.6	4.6	-3.78	0.05	1.3	3.5
33	0.2	2.3	-2.6	0.3	-2.9	2.4	-0.40	0.05	0.5	9.0
34	1.5	1.5	0.1	0.3	-1.4	1.5	-0.66	0.05	0.5	6.4
35	9.8	1.7	-0.4	0.3	-10.2	1.7	-1.71	0.05	0.9	13.9
36	0.8	7.3	-2.4	0.3	-3.2	7.3	-0.16	0.05	0.2	1.3
37	-12.0	2.2	-3.9	0.3	8.0	2.3	2.74	0.05	0.8	1.1
38	15.6	2.5	13.6	0.3	-2.0	2.5	2.35	0.05	0.8	0.8
39	19.0	7.7	21.0	0.3	2.0	7.7	7.43	0.05	1.5	1.1
40	27.5	1.6	26.5	0.3	-1.0	1.6	0.88	0.05	0.8	1.1
41	41.0	6.0	21.1	0.3	-20.0	6.0	3.53	0.05	1.8	1.4
42	-24.8	12.0	-9.4	0.3	15.4	12.0	6.36	0.05	2.2	1.6
43	-18.7	3.3	-15.9	0.3	2.8	3.3	2.70	0.05	1.2	4.6
44	-7.3	26.0	-10.1	0.3	-2.8	26.0	-6.56	0.05	2.8	8.8
46	13.8	6.5	-1.9	0.3	-15.7	6.5	-0.12	0.05	0.4	12.9
47	15.0	1.5	12.3	0.3	-2.7	1.5	1.39	0.05	0.8	1.3
48	28.7	2.6	24.8	0.3	-3.9	2.6	0.36	0.05	0.7	1.4
49	26.7	1.6	23.2	0.3	-3.5	1.6	-1.29	0.05	0.9	0.8
50	-4.3	11.0	7.5	0.3	11.8	11.0	-9.02	0.05	1.3	0.9
52	7.0	10.0	-2.3	0.3	-9.3	10.0	5.25	0.05	2.0	2.1
54	-6.8	2.4	-4.9	0.3	1.9	2.4	-2.71	0.05	0.6	2.1
55	-6.8	14.0	-4.7	0.3	2.1	14.0	-15.93	0.06	1.3	1.1
56	-9.2	13.0	-3.9	0.3	5.3	13.0	-8.03	0.05	1.7	1.1

Values of  $D_{\text{NH}}^{\text{A}}$ , corresponding to dipolar couplings of the (observed) folded state were obtained using the IPAP method [Ottiger, M., Delaglio, F. & Bax, A. (1998) *J. Magn. Reson.* **131**, 373-8], while dipolar coupling values of the unfolded (invisible, excited) state,

$D_{\text{NH}}^{\text{B}} = D_{\text{NH}}^{\text{A}} - D_{\text{NH}}$  were calculated from measured  $D_{\text{NH}}$  values obtained from fits of relaxation dispersion data recorded at 500 and 800 MHz using the pulse sequences of Fig. 2 and SI Fig. 5. Data for which  $\nu_{\text{N}} < 0.2$  ppm, reduced  $\chi^2 > 2.0$  or errors in  $D_{\text{NH}} > 5$  Hz are not included (gray) in Fig. 4c. Values of  $R_{\text{ZZ}} - R_1$  have been measured experimentally and correspond to the difference in relaxation rates between  $2S_{\text{Z}}I_{\text{Z}}$  ( $R_{\text{ZZ}}$ ) and  $S_{\text{Z}}$  ( $R_1$ ). Note that the cross-relaxation rate between TROSY and anti-TROSY magnetization components due to  $^1\text{H}$  spins is given by  $0.5(R_{\text{ZZ}} - R_1)$ . Fitted values of  $k_{\text{ex}} = 470 \pm 10 \text{ s}^{-1}$  and  $p_{\text{B}} = 4.2 \pm 0.05\%$  (population of unfolded state) were obtained.