Table 2. Extracted values of D_{NH}, D_{NH}^{A}, D_{NH}^{B}, Δ_N and reduced χ^2 obtained from fits of relaxation dispersion data recorded on the Abp1p SH3 domain with 6.8% bound Ark1p peptide, ~ 25 mg/ml Pf1 phage, 25°C.

Residue	D_{NH}(Hz)	err	D_{NH}^{A}	err	D_{NH}^{B}	err	Δ_N(ppm)	err	χ^2_{red}	$R_{zz}-R_1$
3	−8.5	4.1	5.4	0.3	13.9	4.1	−0.17	0.05	0.5	1.0
5	−6.7	1.5	3.8	0.3	10.5	1.5	−0.45	0.05	0.5	1.3
8	6.7	1.5	−4.1	0.3	−10.8	1.5	0.88	0.05	0.7	1.5
9	11.1	1.7	−5.9	0.3	−17.0	1.7	−0.32	0.05	0.5	0.9
10	3.7	1.5	−4.4	0.3	−8.1	1.5	−0.59	0.05	0.5	1.1
11	3.4	1.5	−4.5	0.3	−7.9	1.6	0.34	0.05	0.5	0.8
12	0.1	2.0	−0.2	0.3	−0.3	2.1	0.28	0.05	0.4	1.2
13	−5.3	1.8	4.4	0.3	9.7	1.8	0.30	0.05	0.6	1.9
14	−2.2	1.5	2.3	0.3	4.5	1.5	1.11	0.05	0.7	1.5
15*	−9.1	2.7	3.0	0.3	12.1	2.7	2.00	0.05	1.4	11.0
16	2.3	1.5	−2.6	0.3	−4.9	1.5	0.71	0.05	0.8	3.0
17	−4.9	1.5	2.0	0.3	7.0	1.5	1.03	0.05	0.5	1.1
18	−7.6	1.5	6.1	0.3	13.7	1.5	0.49	0.05	0.7	0.6
19	3.5	1.5	−6.9	0.3	−10.3	1.5	0.40	0.05	0.4	1.0
23	−5.4	4.6	6.1	0.3	11.5	4.6	0.13	0.05	0.5	1.0
26	−1.9	4.0	3.8	0.3	5.7	4.0	0.16	0.05	0.6	0.9
29	−0.8	3.3	4.9	0.3	5.7	3.3	0.17	0.05	0.5	0.9
30	−3.0	1.5	0.0	0.3	3.0	1.5	0.43	0.05	0.6	0.9
31	3.9	2.3	−4.3	0.3	−8.2	2.3	−2.00	0.05	1.4	3.5
33	3.9	1.7	6.0	0.3	2.1	1.7	−1.76	0.05	1.1	1.2
34	−0.7	3.8	6.0	0.3	6.7	3.8	−3.70	0.05	1.4	2.1
35	−1.1	1.5	4.4	0.3	5.5	1.5	0.93	0.05	0.7	2.6
36	0.6	1.6	5.0	0.3	4.3	1.6	0.49	0.05	0.7	2.2
37	6.2	2.7	1.2	0.3	−5.0	2.7	1.70	0.05	1.2	1.6
49	−6.8	1.5	2.1	0.3	8.9	1.5	0.92	0.05	1.2	1.1
52	1.5	1.5	6.7	0.3	5.2	1.5	1.30	0.05	1.0	1.6
53	−2.2	1.5	3.7	0.3	6.0	1.5	−0.86	0.05	1.0	1.6

Values of D_{NH}^{A}, corresponding to dipolar couplings of the (observed) apo 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 invisible bound state, $D_{NH}^{B} = D_{NH}^{A} - D_{NH}$ were calculated from measured D_{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 $\Delta_N < 0.2$ ppm or reduced $\chi^2 > 2.0$ are not included (gray) in Fig. 3*e*. Values of $R_{zz}-R_1$ have been measured experimentally and correspond to the difference in relaxation rates between $2S_Z I_Z$ (R_{ZZ}) and S_Z (R_1). Note that the cross-relaxation rate between TROSY and anti-TROSY magnetization components due to

external ^{1}H spins is given by $0.5(R_{ZZ}\text{-}R_1)$. Fitted values of $k_{on}[L]+k_{off} = 374\pm5$ s^{-1}

($P + L \underset{k_{off}}{\overset{k_{on}}{\rightleftharpoons}} PL$) and $p_B = 6.8\pm0.05\%$ (population of bound state) were obtained.

*This residue is the largest outlier in the correlation plot of Fig. 3e (see main text) that may

reflect the fact that the ^{1}H spin flip rate is 3-fold faster than for all other residues.