

SUPPORTING INFORMATION

Table S1. Fit parameters for GCN4-58^a.

Residue (population, %)	Log ₁₀ \bar{R} (τ , nsec)
6	8.60 (0.42)
10	8.40 (0.66)
14 (comp 1, 46%)	8.45 (0.59)
14 (comp 2, 54%)	7.78 (2.77)
17 (comp 1, 36%)	8.36 (0.73)
17 (comp 2, 64%) ^b	7.87 (2.25)
18 (comp 1, 29%)	8.39 (0.68)
18 (comp 2, 71%) ^b	7.94 (1.91)
20 (comp 1, 47%)	8.50 (0.53)
20 (comp 2, 53%) ^b	7.94 (1.91)
25 (comp 1, 32%)	8.13 (1.24)
25 (comp 2, 68%)	7.81 (2.58)
26 (comp 1, 21%)	8.12 (1.26)
26 (comp 2, 79%) ^b	7.73 (3.10)
29	7.75 (2.90)
32 (comp 1, 25%)	8.13 (1.24)
32 (comp 2, 75%)	7.73 (3.10)
35 (comp 1, 24%)	8.00 (1.67)
35 (comp 2, 76%)	7.66 (3.65)
39	7.93 (1.96)
43	7.79 (2.70)
46 (comp 1, 32%)	8.05 (1.49)
46 (comp 2, 68%)	7.54 (4.80)

^a For 6 – 20, N was fixed at 1; for 25-43, N was fixed at 0.3 (see Methods)

^b A log₁₀ Heisenberg exchange term was used for sites 17, 18, 20, and 26 equal to 7.24, 6.96, 7.20, and 6.96, respectively.

Table S2. Fit parameters for GCN4-58 bound to 2AP-1^a.

Residue (population, %)	Log₁₀ \bar{R} (τ, nsec)	C₂₀ (S₂₀)
6	8.03 (1.55)	1.26 (0.28)
7 (comp 1, 60%)	8.07 (1.40)	1.16 (0.26)
7 (comp 2, 40%)	7.55 (4.70)	0 (0)
10	7.95 (1.87)	1.29 (0.29)
17 (comp 1, 92%)	8.01 (1.60)	1.77 (0.39)
17 (comp 2, 8%)	6.77 (2.80)	0 (0)
18 ^b	8.00 (1.67)	1.90 (0.42)
25 (comp 1, 46%)	7.98 (1.75)	1.24 (0.28)
25 (comp 2, 54%)	7.44 (6.10)	0 (0)
29 (comp 1, 92%)	7.85 (2.35)	1.40 (0.32)
29 (comp 2, 8%)	7.59 (4.18)	0 (0)
32 (comp 1, 52%)	7.99 (1.70)	1.29 (0.29)
32 (comp 2, 48%)	7.53 (4.80)	0
39	7.94 (1.90)	1.28 (0.28)
43	7.86 (2.30)	0.93 (0.21)
46 (comp 1, 52%)	7.96 (1.83)	0 (0)
46 (comp 2, 48%)	7.54 (4.70)	0 (0)

^a N=0.3 for all fits (see Methods)^b C₂₂=-1.1**Table S3.** Fit parameters for R1(b).

Residue	Log₁₀ \bar{R} (τ, nsec)	C₂₀ (S₂₀)	Log₁₀ N
29R1(b) free	7.69 (3.40)	1.27 (0.28)	-.20
29R1(b) bound	7.67 (3.40)	1.50 (0.33)	-.20
35R1(b) free ^a	7.70 (3.30)	2.15 (0.47)	-.34

^a C₂₂=-1.0