Supporting Information for World Wide Web Edition

Table 1: Estimated Conformational Exchange Rates

Site	K_{off} (s^{-1})
I	268 > x > 104
II	780 > x > 91
III	< 82
IV	< 60

Individual estimates (s^{-1}): K21 > 184, D22 < 315, G23 > 180, G25 > 177, T26 > 245, I27 = 725, T28 < 268, T29 < 340, G33 > 152, V35 > 213, A57 > 304, D58 > 175, G59 > 146, N60 < 780, G61 > 155, T62 > 91, I63 > 230, D64 > 108, F65 > 94, F68 > 165, L85 < 145, D93 < 282, R94 < 287, D95 < 244, G96 < 131, G98 < 528, I100 < 637, S101 < 264, L105 < 209, M109 < 100, T110 < 82, G113 < 116, L116 < 189, T117 < 99, A128 < 240, I130 < 259, G132 < 330, G134 < 490, I136 < 161, N137 < 387, Y138 < 547, E139 < 355, F141 < 489, K148 < 60.

Table 2:
NMR assignments for PCaM under apo conditions (*see Materials and Methods*).

Residue	NH	N	CA	CB	CO
A1					
E2					
Q3	8.35	119.7	55.52	30.18	175.66
L4	8.26	122	54.42	43.58	177.5
T5	8.78	112.83	60.48	71.16	175.49
E6	9.06	120.5	60.04	29.29	179.51
E7	8.76	119.5	60.29	28.96	179.28
Q8	7.75	120.5	58.76	29.19	177.68
I9	8.26	118.5	66.79	38.1	177.72
A10	7.97	120.7	55.48	18.06	180.63
E11	7.78	119.98	59.44	29.5	180.28
F12	8.78	120.5	58.51	37.16	178.2
K13	9.21	121.5	59.99	32.32	178.68
E14	7.95	118.7	59.2	29.55	178.7
A15	7.69	120.72	55	19.31	179.03
F16	8.46	118.5	61.98	40.87	177.22
A17	8.41	117.5	54.59	18.51	179.95
L18	7.36	117.03	57.04	41.85	178.09
F19	7.51	115.3	58.47	39.94	174.69
D20	7.43	123.2	52.67	39.93	177.09
K21	8.07	123.7	58.51	32.55	177.97
D22	8.75	117.27	54.56	41.48	176.87
G23	8.06	110.36	46.9		175.29
D24	8.86	121	53.99	41.67	177.42
G25	10.31	112.33	46.03		174.32
T26	7.63	110.1	59.95	73.26	172.88
I27	8.3	110.6	59.41	41.52	176.14
T28	8.36	111.1	60.76	70.65	176.98
T29	8.31	112.58	65.12	67.36	177.22
K30	7.62	119	58.32	32.36	177.93
E31	7.57	117.52	55.93	29.87	176.24
L32	7.39	121.2	58.71	41.85	178.16
G33	8.86	105.4	48.27		174.96
T34	7.55	118.7	66.25	68.32	177.27
V35	8.03	122.7	66.39	31.3	179.32
M36	8.5	119	60.85	32.3	178.95
R37	8.58	119.5	59.31	30.18	181.34
S38	8.12	119	61.71	62.87	174.99
L39	7.38	121.47	54.55	43.04	177.33
G40	7.94	107.15	45.86		174.54
Q41	7.83	118	54.11	30.06	174.17
N42	8.7	117.03	51.23	39.45	176.82
P43			62.43	31.96	177.66
T44	8.8	113.6	60.77	70.94	175.17
E45	8.89	120.5	60.11	29.23	179.22

A46	8.35	121.47	55.12	18.38	180.22
E47	7.78	119.2	58.89	29.99	179.92
L48	8.46	120.2	58.21	41.93	178.39
Q49	8.1	118	58.79	28.16	178.31
D50	7.84	119.25	57.64	40.49	179.12
M51	8.1	119.25	59.95	33.84	178.14
I52	8.48	119.5	65.83	37.98	177.47
N53	8.18	117	55.32	38.27	176.77
E54	7.66	117.8	58.7	30.28	178.29
V55	7.84	112.3	62.51	32.72	175.8
D56	8.57	122.4	53.25	40.78	177.2
A57	8.17	125.66	55.21	18.95	179.39
D58	8.49	114.31	54.65	41.08	177.09
G59	7.9	109.1	47.02		175.81
N60	9.62	120.67	54.74	39.59	177.46
G61	10.44	110.5	47.1		174.87
T62	7.5	110.61	59.6	72.71	172.8
I63	8.93	118.75	60.16	41.77	174.43
D64	8.52	124.67	52.11	42.21	176.28
F65	8.67	118.57	63.16	36.55	173.71
P66			66.63	30.59	180.14
E67	8.18	118.2	59.46	29.43	179.15
F68	8.55	122.4	61.76	39.58	176.61
L69	8.66	119	57.7	41.23	179.05
S70	7.66	114.06	61.47	62.62	177.1
L71	7.74	122.45	57.84	41.84	179.13
M72	7.94	117.03	55.93	31.02	178.78
A73	8.39	121	54.87	18.09	179.8
R74	7.52	117.03	58.91	30.48	178.33
K75	7.89	118.7	56.61	31.35	178.17
M76	8.12	118.26	56.89	32.93	177.07
K77	7.8	120.48	57.26	32.93	177.28
E78	8.25	121.46	57.35	30.23	177.06
Q79	8.25	120.2	56.61	29.67	176.16
D80	8.44	122.2	54.93	41.41	176.75
S81	8.43	116.77	59.57	63.88	175.51
E82	8.65	122.95	59.18	29.87	177.41
E83	8.37	119	59.94	29.79	178.45
E84	8.14	119	59.05	29.79	179.09
L85	7.98	121.2	57.87	42.6	178.11
I86	8.3	118.26	65.36	37.37	178.11
E87	8.03	117.5	59.2	29.35	178.45
A88	7.62	121.2	54.59	18.24	179.72
F89	7.73	114.8	59.71	39.45	177.79
K90	8.25	118.26	59.2	32.94	178.55
V91	7.01	116.8	64.83	32.02	176.72
F92	7.48	116.3	58.08	39.53	174.96
D93	7.81	121.7	52.48	40.12	176.59
R94	8.32	124.4	58.31	30.44	178

D95	8.7	116.8	54.65	41.41	176.81
G96	8.03	110.36	47		175.11
N97	8.89	118	52.47	39.21	176.36
G98	9.84	111	46.12		173.24
L99	7.63	119.5	53.3	44.94	176.53
I100	8.72	115	59.5	41.97	174.57
S101	8.82	119	57.79	65.16	175.56
A102	8.59	124.7	55.54	18.06	178.96
A103	8.33	118.5	55	18.38	181.1
E104	7.67	119.6	58.66	28.89	177.82
L105	8.2	121.47	59.09	41.86	179.06
R106	8.29	117.76	60.15	30.06	178.54
H107	7.74	119	59.3	29.63	177.63
V108	8.03	120.2	66.42	31.97	178.61
M109	8.19	115.8	57.07	31.55	178.81
T110	8.02	113.08	65.04	69.22	175.83
N111	7.73	119.5	54.59	39.36	175.89
L112	7.8	119.98	55.45	43	177.74
G113	8.15	107.65	46.16		174.77
E114	8.12	119.7	56.64	29.82	176.51
K115	8.41	120.7	56.99	32.67	176.3
L116	7.8	121.2	54.14	44.61	177.55
T117	9.02	113.81	61.04	70.91	175.46
D118	8.8	121.22	57.81	40.27	178.18
D119	8.32	117.3	57.16	40.39	178.8
E120	7.78	120.7	59.27	30.41	179.39
V121	8.27	120.7	67.2	31.79	177.66
D122	8.41	119.59	57.89	40.61	179.16
E123	7.84	119.7	59.27	29.55	178.32
M124	7.95	119.15	59.27	34.45	178.77
I125	8.61	119.2	66.17	37.78	177.32
R126	7.8	118.26			179.26
E127	7.82	117.03	59.4	30.26	177.1
A128	7.83	120.7	52.32	19.61	176.3
D129	7.84	121	53.81	40.3	177.2
I130	8.4	123.2	62.57	39.08	177.2
D131	8.55	120.72	53.72	41.51	177.29
G132	8.26	109.9	46.56		174.41
D133	8.48	120.23	53.31	41.78	176.43
G134	9.51	110.86			174.55
H135	8.1	119.7	55.32	29.9	172.63
I136	9.14	122	59.86	39.6	175.2
N137	8.72	125.2	52.48	37.92	175.65
Y138	7.59	125.2	59.65	37.07	176.4
E139	8.38	125.4	60.7	28.91	179.15
E140	7.77	119.3	58.56	29.39	178.54
F141	8.1	120.5	62.11	39.84	177.01
V142	8.5	118.4	67.23	31.62	177.33
R143	7.8	118.7	59.64	29.79	179.18

M144	7.87	117.5	58.24	32.93	178.11
M145	8.06	119.5	58.69	33.34	178.1
V146	8.02	113.6	63.1	32.08	176.6
S147	7.68	116.8	58.68	63.87	173.59
K148	7.54	128.1	57.96	33.51	181.41

Table 3: NMR assignments for Ca^{2+}-saturated PCaM (see *Materials and Methods*).

Residue	NH	N	CA	CB	CO
A1					
E2					176.06
Q3	8.468	121.36	55.51	29.79	175.6
L4	8.294	123.28	54.35	43.79	177.69
T5	8.785	113.29	60.41	68.12	175.45
E6	9.08	120.3	60.13	29.24	179.52
E7	8.753	119.5	60.11	29.1	
Q8	7.77	120.45	58.68	29.28	175.34
I9	8.413	119.5	66.34	37.96	177.72
A10	8.02	121.15	55.44	17.89	
E11	7.797	119.5	58.26	29.26	180.53
F12	8.61	120.07	59.31	37.94	178.65
K13	9.201	123.55	60.38	32.17	179.39
E14	7.76	119.8	59.12	29.25	179.06
A15	7.99	122.2	55.2	18.07	178.84
F16	8.798	119.23	62.02	39.76	176.58
A17	7.714	118.02	54.46	18.28	179.24
L18	7.46	117.4	57.03	41.73	178.35
F19	7.51	114.9	59.3	40.25	176.54
D20	7.816	119.3	52.51	38.7	177.26
K21	7.701	124.36	58.7	32.64	178.05
D22	8.12	113.6	52.96	39.59	177.59
G23	7.7	109	47.18		175.08
D24	8.357	120.34	53.82	40.48	177.4
G25	10.663	113.43	45.56		173.71
T26	8.12	113.3	59.8	69.7	172.962
I27	9.75	126.25	60.03	39.54	175.92
T28	8.487	116.4	59.87	69.01	176.46
T29	8.99	113.3	66.4	67.8	
K30	7.78	120.6	59.2	32.4	179.8
E31	7.622	120.72	58.93	30.11	178.98
L32	8.722	120.72	57.77	42.24	178.81
G33	8.556	106.14	48.48		176.49
T34	7.76	117.6	66.8	68.6	177.09
V35	7.605	122.07	66.08	31.32	179.14
M36	8.424	118.15	58.7	31.2	179.17
R37	8.48	119.5	59.2	30.06	181.22
S38	7.93	119	61.7	62.95	174.85
L39	7.365	120.58	54.55	42.04	177.45
G40	7.889	106.81	45.63		174.36
Q41	7.87	118.7	54.46	30.64	174.07
N42	8.722	116.53	51.29	39.24	
P43			62.4	32.08	177.59
T44	8.77	112.8	60.58	68.11	175.45
E45	9.052	120.45	60.16	29.19	178.97
A46	8.297	120.72	55.09	18.26	180.23

E47	7.701	118.42	59.07	29.8	179.42
L48	8.104	122.07	57.98	42.25	
Q49	8.16	118.2	58.56	28.22	178.41
D50	8.057	119.77	57.56	40.33	178.7
M51	7.886	119.5	59.46	33.38	
I52	7.798	118.12	65.13	37.36	177.7
N53	8.427	117.34	55.84	38	177.39
E54	7.621	117.07	58.93	30.38	177.8
V55	7.288	108.67	61.03	32.8	175.97
D56	7.795	121.93	53.92	40.12	176.38
A57	8.549	131.65	54.22	19.48	178.62
D58	8.14	114	52.6	39.7	178.29
G59	7.607	108.67	47.2		
N60	8.06	118.2	52.42	37.85	176.85
G61	10.594	113.56	45.78		173.32
T62	7.682	109.24	59.56	68.89	173.14
I63	8.979	123.28	59.6	39.65	175.3
D64	8.736	127.87	52.09	42.01	176.27
F65	8.858	118.56	63.74	35.99	
P66			66.58	30.58	
E67	8.03	118.2	58.94	29.59	178.73
F68	8.88	123.3	61.38	39.89	176.99
L69	8.42	119	57.83	40.88	179.2
S70	7.457	113.97	61.54	62.73	176.71
L71	7.775	123.15	58.05	41.9	178.43
M72	8.072	116.53	55.92	31.28	
A73	8.153	121.53	54.72	18.22	179.73
R74	7.507	117.34	58.54	30.14	178.27
K75	7.98	118.9	57.29	31.91	178.27
M76	8.09	118.7	57.18	32.84	
K77	7.78	120.3	57.18	32.73	177.45
E78	8.18	120.9	57.5	29.94	177.21
Q79	8.201	120.45	56.58	29.43	176.28
D80	8.389	121.86	55.05	41.19	176.84
S81	8.351	116.46	59.21	63.88	175.37
E82	8.509	123.15	58.54	29.76	175.07
E83	8.396	119.23	59.3	29.4	178.46
E84	8.232	119	58.96	29.86	180.25
L85	8.27	120.6	57.93	42.19	178.83
I86	8.3	121.7	65.01	38.19	178.45
E87	7.93	118.4	59.03	29.13	179.44
A88	8.09	122.2	55.21	18.22	179.5
F89	8.65	120.1	62.29	39.31	176.88
K90	7.85	116	59.38	32.8	178.24
V91	7.361	118.42	65.83	31.65	177.31
F92	7.221	116.13	60.48	40.82	
D93	7.891	116.13	52.24	38.59	177.4
R94	7.747	124.93	59.26	30.75	178
D95	8.212	114.1	53.02	39.72	177.96

G96	7.769	108.7	47.25		175.08
N97	8.207	119.23	52.59	37.86	176.14
G98	10.895	113.7	45.55		172.95
L99	7.487	119.23	52.74	46.5	175.64
I100	9.993	128.14	61.25	38.63	175.92
S101	9.32	124.2	55.7	67.1	174.99
A102	9.17	123.3	55.8	18.1	
A103	8.22	118.4	55.15	18.45	181.64
E104	7.882	120.51	59.59	29.06	178.76
L105	8.617	120.58	58.61	42.47	178.56
R106	8.622	117.07	59.84	30.68	
H107	7.977	119.23	59.92	30.95	
V108	8.11	118.7	66.34	31.96	178.11
M109	8.39	116.7	57.9	31.28	178.6
T110	8.08	114.7	65.7	69.00	177.58
N111	7.87	122.07	55.65	38.35	
L112	7.927	119.23	55.37	42.04	177.36
G113	7.847	106.78	45.47		174.36
E114	7.939	120.45	55.31	30.38	175.59
K115	8.547	123.82	55.63	32.07	175.72
L116	8.101	124.93	54.18	44.75	178.05
T117	9.218	114.78	60.7	68.01	175.53
D118	8.864	120.9	58.06	39.77	178.61
D119	8.3	117.9	57.17	40.35	178.65
E120	7.733	120.72	59.3	30.46	
V121	8.127	121.53	66.98	31.49	
D122	8.01	119.5	57.64	40.45	176.98
E123	8.01	119.8	59.23	29.47	
M124	7.82	119.8	59.37	33.2	
I125	8	118.2	64.73	36.94	178.45
R126	8.24	118.45	59.72	30.27	179.35
E127	7.906	115.59	58.45	29.78	177.2
A128	7.35	118.7	52.1	21.4	
D129	8.05	118.2	54.15	40.3	176.05
I130	8.41	127.8	62.9	38.3	177.77
D131	8.25	117.1	52.6	39.9	178.29
G132	7.6	109	47.64		175.32
D133	8.39	121.2	53.6	40.5	178.95
G134	10.49	115.2	46.1		172.27
H135	8.297	116.29	54.21	34.97	174.56
I136	9.363	125.17	59.63	39.34	176.06
N137	9.493	129.22	51.18	38.03	175.34
Y138	8.684	119.16	63.05	37.89	
E139	8.11	118.39	60.2	29.15	180.47
E140	8.801	119.5	58.26	30.04	179.22
F141	9.076	124.76	61.45	39.91	
V142	8.438	119.5	67.21	31.66	177.95
R143	7.243	116.53	58.92	29.73	
M144	7.83	118.45	57.98	33.45	

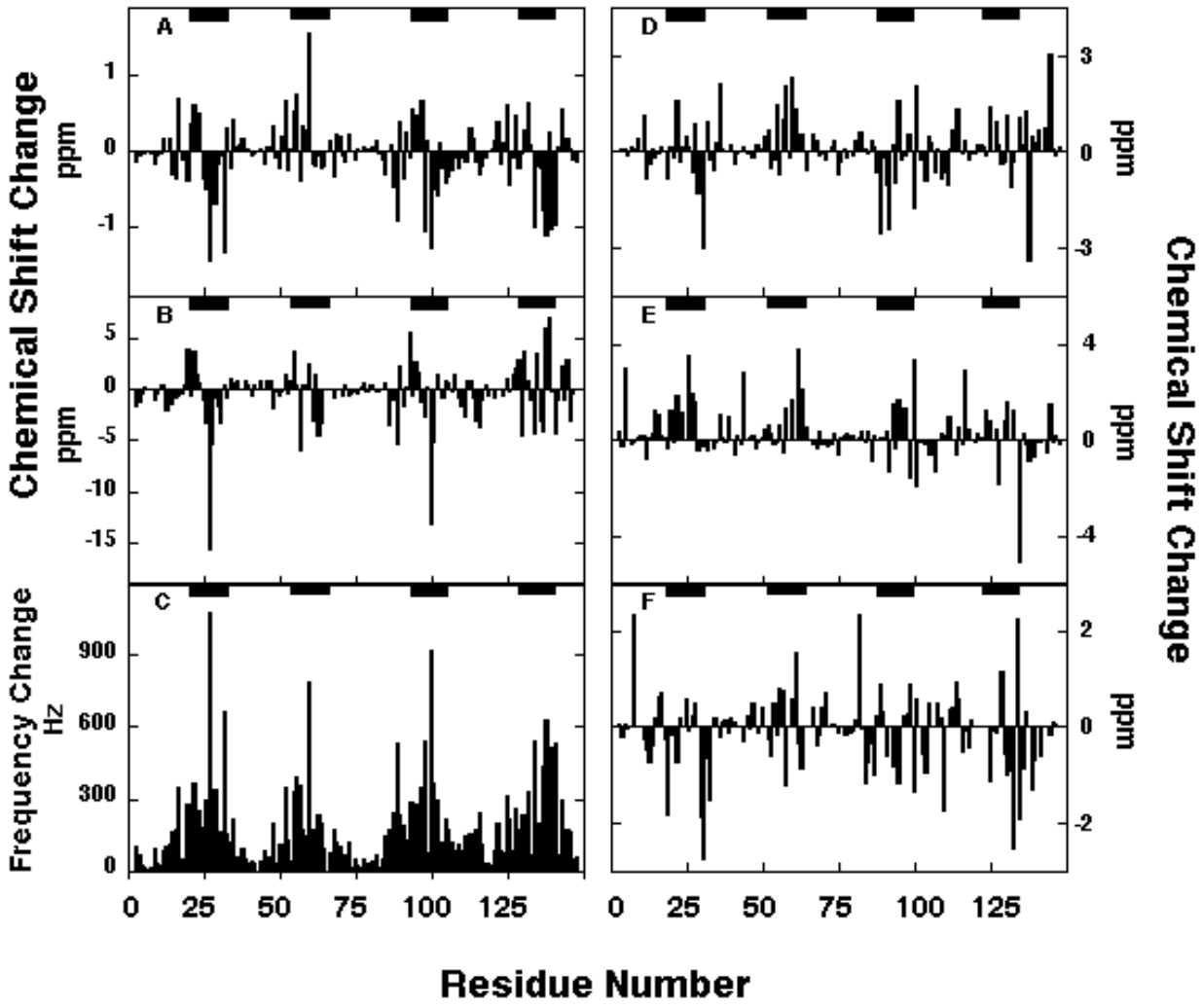
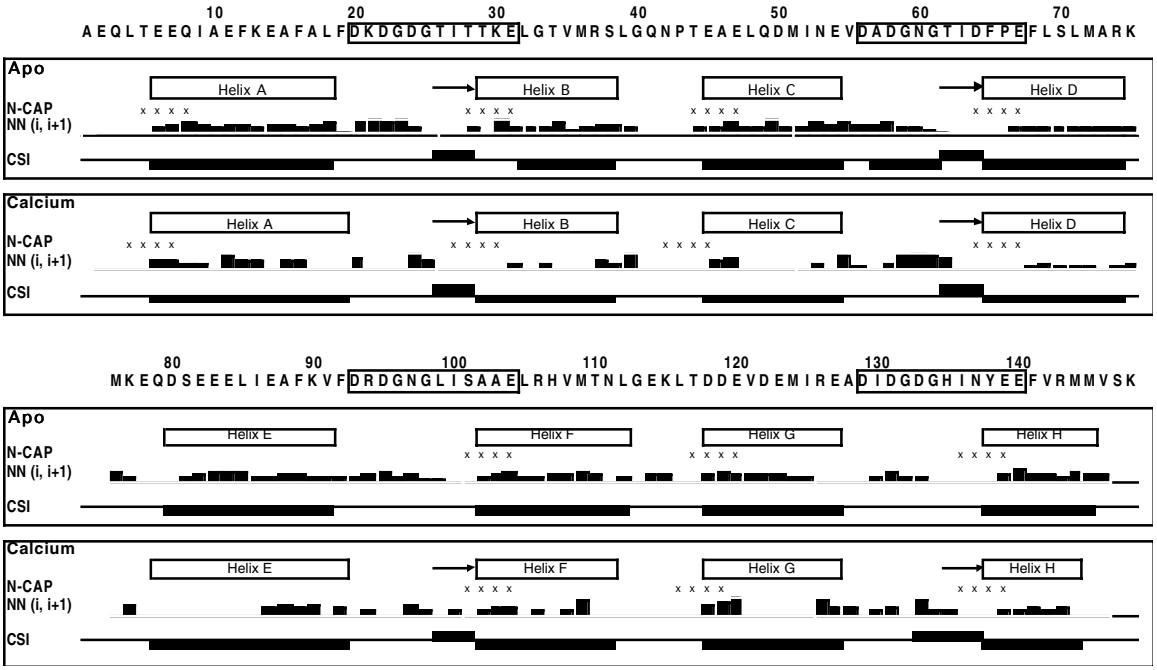
M145	7.88	116.56	55.62	31.83	178.26
V146	7.97	116.7	63.04	32.13	176.49
S147	7.77	117.2	58.77	63.7	173.54
K148	7.66	128.1	57.83	33.64	

Figure Legends

Figure 1: Calcium-dependent changes in chemical shifts ($\delta_{apo} - \delta_{calcium}$) of atoms on the protein backbone of PCaM. (A) amide proton, (B) amide nitrogen, (C) weighted average of the proton and nitrogen frequency differences at 500 MHz, (D) alpha carbon, (E) beta carbon, (F) carbonyl carbon. Black bars indicate calcium-binding sites.

Figure 2: Comparison of predicted secondary structures of apo and calcium-saturated PCaM based on NOE and chemical shift measurements. The amino acid sequence is shown on top, with the 12-residue calcium-binding sites in boxes. The predicted secondary structure is indicated by boxes (helix) and arrows (beta strand) underneath the amino acid sequence. Data used to make the prediction of structure is shown below the prediction. N-CAP refers to the N-terminal helix-capping motif (*44*) and is indicated by xxxx. NN (i, i+1) refers to the amide proton NOEs between i and i+1 residues. CSI refers to the chemical shift index described by Sykes et al. ((*45*); http://www.pence.ca).

Supporting Information
Figure 1
Jaren, Kranz et al.



Supporting Information
Figure 2
Jaren, Kranz et al.