Table S1. Crystallographic statistics, Related to Figures 1, 2, and 3

	***Ab*Cap4 Apo**	***Ab*Cap4–2′3′3′-cAAA**	***Ab*Cap4–3′3′3′-cAAA**	***Ab*Cap4–2′3′3′-cAAA** (SeMet)
Data Collection				
Resolution (Å)[a]	39.60–2.60 (2.64–2.60)	49.21–2.10 (2.14–2.10)	38.80–2.40 (2.44–2.40)	49.46–2.45 (2.49–2.45)
Wavelength (Å)	0.97918	0.97918	0.97918	0.97918
Space group	P 1 2_1 1	P 1 2_1 1	P 1 2_1 1	P 1 2_1 1
Unit cell: a, b, c (Å)	99.72 111.38 173.30	106.58, 111.46, 164.83	106.50 111.18 163.75	106.37, 112.23, 163.35
Unit cell: α, β, γ (°)	90.00 103.06 90.00	90.00, 100.21, 90.00	90.00 100.36 90.00	90.00, 100.03, 90.00
Molecules per ASU	6	6	6	6
Total reflections	335131	1567459	519258	3953693
Unique reflections	100775	217277	145531	138853
Completeness (%)[a]	88.7 (48.8)	98.3 (95.5)	99.0 (86.2)	99.9 (99.9)
Multiplicity[a]	3.3 (2.2)	7.2 (7.1)	3.6 (3.1)	28.5 (27.8)
$I/\sigma I$[a]	12.9 (2.3)	12.4 (1.6)	8.4 (1.0)	14.1 (1.3)
CC(1/2)[b] (%)[a]	99.7 (80.8)	99.8 (55.0)	99.6 (42.8)	99.9 (66.4)
Rpim[c] (%)[a]	3.8 (29.9)	4.2 (81.1)	5.8 (88.1)	3.4 (63.3)
Sites				36
Refinement				
Resolution (Å)	39.60–2.60	49.21–2.10	38.62–2.40	
Free reflections	1993	2000	2000	
R-factor / R-free	19.2 / 23.1	18.5 / 21.9	20.8 / 24.0	
Bond distance (RMS Å)	0.004	0.003	0.002	
Bond angles (RMS °)	0.59	0.53	0.50	
Structure/Stereochemistry				
No. atoms: protein	20800	21078	20894	
No. atoms: ligand	30 (SO_4)	370 (2′3′3′-cAAA, SO_4)	426 (3′3′3′-cAAA, SO_4)	
No. atoms: solvent	317	1490	492	
Average B-factor: protein	51.64	54.03	65.48	
Average B-factor: ligand	58.79	69.46	64.86	
Average B-factor: water	41.14	50.34	52.51	
Ramachandran plot: favored	97.46%	97.84%	96.73%	
Ramachandran plot: allowed	2.50%	2.16%	3.19%	
Ramachandran plot: outliers	0.04%	0%	0.08%	
Rotamer outliers	1.81%	0.68%	0.86%	
MolProbity[d] score	1.48	1.29	1.29	
Protein Data Bank ID	6WAM	6VM6	6WAN	

[a] Highest resolution shell values in parenthesis
[b] (Karplus and Diederichs, 2012)
[c] (Weiss, 2001)
[d] (Chen et al., 2010)

Table S1. Crystallographic Statistics, Related to Figures 1, 2, and 3

	***Mo*Cap4 Apo**	***Mo*Cap4** (SeMet)
Data Collection		
Resolution (Å)[a]	49.36–2.34 (2.43–2.34)	49.30–2.80 (2.95–2.80)
Wavelength (Å)	0.99998	0.97949
Space group	P 3_2 2 1	P 3_2 2 1
Unit cell: a, b, c (Å)	157.59, 157.59, 63.32	157.82, 157.82, 63.16
Unit cell: α, β, γ (°)	90.0, 90.0, 120.0	90.0, 90.0, 120.0
Molecules per ASU	1	1
Total reflections	423237	2521929
Unique reflections	37860	22581
Completeness (%)[a]	99.2 (92.4)	99.8 (98.8)
Multiplicity[a]	11.2 (10.6)	111.7 (108.1)
$I/\sigma I$[a]	15.3 (0.9)	19.8 (1.7)
CC(1/2)[b] (%)[a]	99.9 (41.3)	100 (69.5)
Rpim[c] (%)[a]	3.2 (89.5)	3.2 (60.8)
Sites		5
Refinement		
Resolution (Å)	49.36–2.35	
Free reflections	2004	
R-factor / R-free	20.7 / 23.4	
Bond distance (RMS Å)	0.006	
Bond angles (RMS °)	0.710	
Structure/Stereochemistry		
No. atoms: protein	3385	
No. atoms: ligand	1 (Mg)	
No. atoms: solvent	67	
Average B-factor: protein	83.51	
Average B-factor: ligand	99.78	
Average B-factor: water	65.29	
Ramachandran plot: favored	97.07%	
Ramachandran plot: allowed	2.93%	
Ramachandran plot: outliers	0.0%	
Rotamer outliers	0.0%	
MolProbity[d] score	1.46	
Protein Data Bank ID	6VM5	

[a] Highest resolution shell values in parenthesis
[b] (Karplus and Diederichs, 2012)
[c] (Weiss, 2001)
[d] (Chen et al., 2010)