Experimental section

Synthesis and observation of CdSe nanoparticles: 50 mL solution of 0.4 mg/mL apo-r*Li*Dps (recombinant apo-*Li*Dps; r*Li*Dps), 1mM cadmium acetate, 40 mM ammonium acetate, 5mM ammonium was first prepared and the stable cadmium (II) tetraammine ion was formed. Then selenourea was added in two ways: (1) 2.5 mL of 100 mM selenourea was added to the reaction solution; (2) 500 μL of 100 mM selenourea was added to the reaction solution every 25 sec, 5 times. The reaction solutions were left for 14 h at 23 °C. The solutions after 15min, 1h and 14h incubation showed nearly the same core formation ratio. A little precipitate resulted from both methods (1) and (2). On the other hand, a reaction solution without apo-r*Li*Dps showed heavy brown bulk precipitation. It was found that repetitive additions of selenourea showed less precipitation. A small aliquot of the obtained solution was observed by transmission electron microscopy (TEM) with aurothioglucose staining, which was experimentally proven not to stain the r*Li*Dps cavity.[7a]

Purification: The small amount of bulk precipitates in 50 mL reaction solutions were removed by centrifugation at 9000 g for 15 min and the supernatant was filtrated by a 0.22 μm filter. A small aliquot (3 μL) of the obtained solution was observed by TEM with aurothioglucose staining (3 μL) and unstaining[28]. For XRD analysis, the filtered 50 mL solution (method (2)) having r*Li*Dps with CdSe NP was concentrated by ultrafilter membrane (Centriprep 50, Amicon) and then purified by chromatography (Sephacryl S-300 columns, GE Health care Bio-Sciences) using 50 mM Tris pH 8.0 elution buffer. The 12 mer r*Li*Dps fractions were concentrated and washed in milliQ water by ultrafilter membrane (Centriprep 50, Amicon). The 12 mer r*Li*Dps solution was centrifuged at 70000 rpm × 1.5 h, (70.2Ti rotor, Beckman) and the obtained pellet was dried under nitrogen gas flow. The dry pellet was crushed into powder.

Sucrose density gradient: Purified r*Li*Dps solutions with CdSe NP by chromatography were concentrated using ultrafilter membrane (Centriprep 50) and applied to a sucrose density gradient (50 %:10 mL, 45 %:10 mL, 40 %: 10mL) at 28 000 rpm × 23 h (SW28

swing rotor, Beckman). 1 mL of 40 % fraction was extracted as the r*Li*Dps with CdSe NPs. The sucrose solution was replaced by milliQ water using ultrafilter membrane (Centriprep 50) and concentrated to 1 mg/mL.

TEM and XRD analysis: All TEM grids were observed by TEM (JEM2200FS, JEOL) at 200 kV with aurothioglucose staining (3 μL) and unstaining.[3a,f] XRD (Rint-TTR III, Rigaku corp rotary Cu anode operated at 50 kV/300mA) analyses were carried out.

Fig. S1

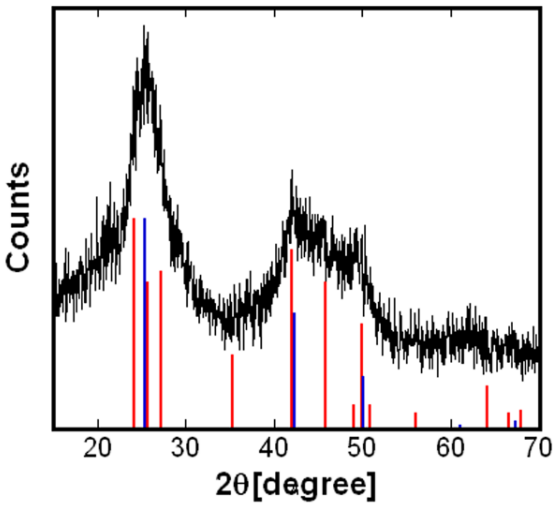

Fig. S1. XRD spectrum from the CdSe-r*Li*Dps. Red lines are peaks of wurtzite (hexagonal). Blue lines are peaks of zinc blend (cubic).

Fig. S2

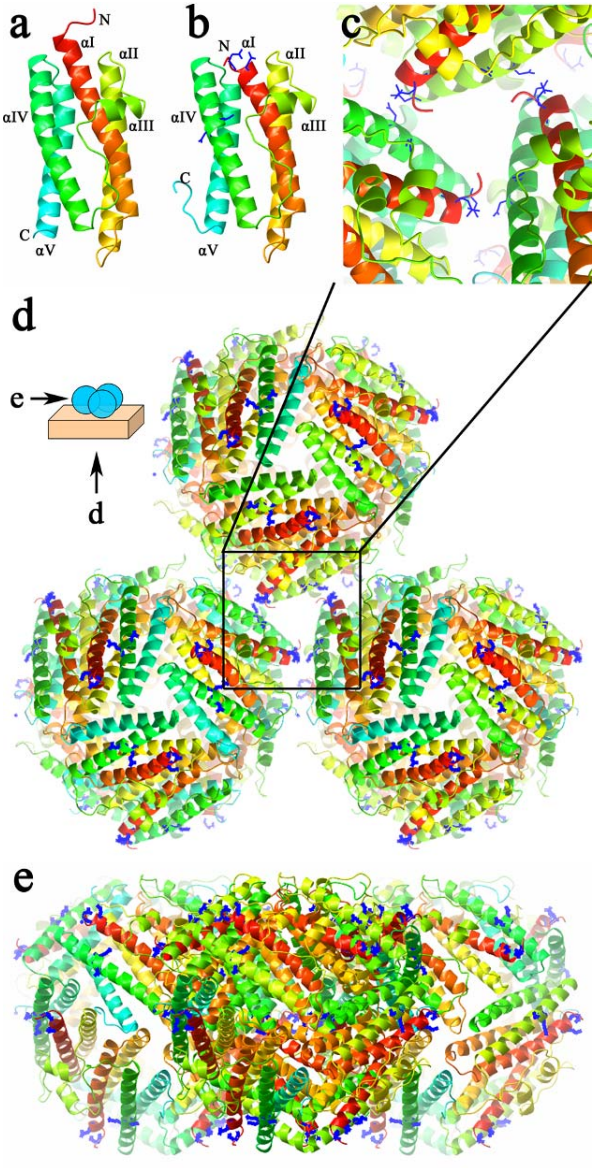

Fig. S2. The ribbon models of (a) The subunit structure of *Bb*Dps (1N1Q) and (b) *Li*Dps (1QGH). Helices αI to αV are shown as red to cyan. The acidic residues (Asp8, Glu11, Asp111 and Glu112) of *Li*Dps are shown as blue sticks. (c) N terminus and αIV gathering around hexagonal packing center of *Li*Dps. (d) A bottom-view from the denatured film or carbon film of a two-dimensional crystal of *Li*Dps. (e) A side-view of

the two-dimensional crystal of (d). The Figures were prepared using CCP4mg (L. Potterton, S. McNicholas, E. Krissinel, J. Gruber, K. Cowtan, P. Emsley, G. N. Murshudov, S. Cohen, A. Perrakis, M. Noble, *Acta Crystallogr., Sect. D: Biol. Crystallogr*. 2004, **D60**, 2288)