Supplementary Information (SI) for Soft Matter.

Supporting Information

Co-Assembly of Nanometer- and Submicrometer-Sized Colloidal Particles into Multi-Component Ordered Superstructures

Javier Fonseca,*[a] Li Jiao,[a] Anna Broto-Ribas[b], Gurvinder Singh,[c] and Angang Dong[d]

[a] Department of Chemical Engineering, Northeastern University, 313 Snell Engineering Center, 360 Huntington Avenue, Boston, Massachusetts 02115-5000, United States.

[b] Departament de Química, Universitat Autònoma de Barcelona, Cerdanyola del Vallès, Barcelona 08193, Spain.

[c] The School of Biomedical Engineering, Faculty of IT and Engineering, Sydney Nano Institute, The University of Sydney, Camperdown, NSW 2008, Australia.

[d] Department of Chemistry and Shanghai Key Laboratory of Molecular Catalysis and Innovative Materials, Fudan University, Shanghai, 200438 China.

*Corresponding author.

E-mail: fonsecagarcia.j@northeastern.edu (Javier Fonseca)

Contents

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles.

Small particles (S) (size in nm)[ϰ]	Large particles (L) (size in nm)[ϰ]	Particle functionalization	Size ratio (γ)[†]	Solvent	Co-assembly approach	Superlattice	Stoichiometry	n_L:n_S	Ref.
Spherical PbSe (6)	Spherical γ-Fe_2O_3 (11)	-	0.55	Dibutyl ether	Solvent evaporation (60 °C)	AlB_2-type superlattice	LS_2	1:2	184
Spherical PbSe (6)	Spherical γ-Fe_2O_3 (11)	-	0.55	Dibutyl ether	Solvent evaporation (60 °C)	$NaZn_{13}$-type superlattice	LS_{13}	1:13	184
Spherical Pd (3)	Spherical PbSe (5.8)	Pd: DDT PbSe: OA	0.627	Toluene	Solvent evaporation (45-50 °C)	(ico-LS_{13}- or) $NaZn_{13}$-type superlattice Cub-LS_{13}-type superlattice	LS_{13}	1:20	29
Spherical CdSe (4.4)	Spherical CdTe (9.1)	CdSe: TOPO and ODA CdTe: ODPA and TOPO	0.57	TCE	Solvent evaporation (55 °C)	AlB_2-type superlattices $NaZn_{13}$-type superlattices Cub-LS_{13}-type superlattice	LS_2 LS_{13} LS_{13}	-	37,38
Spherical CdSe (4.4)	Spherical CdTe (8.1)	CdSe: TOPO and ODA CdTe: ODPA and TOPO	0.63	TCE	Solvent evaporation (55 °C)	$CaCu_5$-type superlattices $NaZn_{13}$-type superlattices Cub-AB_{13}-type superlattices	LS_5 LS_{13} LS_{13}	-	37,38
Spherical CdSe (4.4)	Spherical CdTe (7.0)	CdSe: TOPO and ODA CdTe: ODPA and TOPO	0.71	TCE	Solvent evaporation (55 °C)	$CaCu_5$-type superlattices	LS_5	-	37,38
Spherical CdSe (4.4)	Spherical CdTe (5.9)	CdSe: TOPO and ODA CdTe: ODPA and TOPO	0.81	TCE	Solvent evaporation (55 °C)	$MgZn_2$-type superlattices	LS_2	-	37,38
Spherical Au (5)	Spherical Fe (13)	Au: DDT Fe: OA	0.424	Chloroform	Solvent evaporation (RT)	NaCl-type superlattice	LS	-	190
Spherical Au (5)	Spherical γ-Fe_2O_3 (13.4)	DDT, OA, or TOPO	0.43	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	NaCl-type superlattice AlB_2-type superlattice	LS LS_2	-	31,32
Spherical Pd (3)	Spherical PbSe (7.2)	Pd: DDT PbSe: OA	0.528	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	NaCl-type superlattice	LS	-	32
Spherical Pd (3)	Spherical PbSe (6.2)	DDT, OA, or TOPO	0.59	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	NaCl-type superlattice CuAu-type superlattice Orthorhombic symmetry class AlB_2-type superlattice $MgZn_2$-type superlattice $MgNi_2$-type superlattice Cu_3Au-type superlattice Fe_4C-type superlattice $NaZn_{13}$-type superlattice Cub-LS_{13}-type superlattice	LS LS LS LS_2 LS_2 LS_2 LS_3 LS_4 LS_{13} LS_{13}	-	31,32
Spherical Pd (3)	Spherical PbSe (5.8)	DDT, OA, or TOPO	0.63	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	NaCl-type superlattice AlB_2-type superlattice $MgNi_2$-type superlattice Fe_4C-type superlattice CaB_6-type superlattice $NaZn_{13}$-type superlattice Cub-LS_{13}-type superlattice	LS LS_2 LS_2 LS_4 LS_6 LS_{13} LS_{13}	-	31,32
Spherical Ag (3)	Spherical PbS (6.7)	Ag: DDT PbSe: OA	0.56	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	CuAu-type superlattice	LS	1:3-1:5	32

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Ag (3.4)	Spherical PbSe (5.8)	Ag: DDT PbSe: OA	0.68	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	CuAu-type superlattice	LS	1:3-1:5	32
Spherical Au (5)	Spherical PbSe (7.6)	Au: DDT PbSe: OA	0.70	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	CuAu-type superlattice	LS	1:3-1:5	32
Spherical Au (5)	Spherical PbSe (6.2)	Au: DDT PbSe: OA	0.82	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	CuAu-type superlattice	LS	1:3-1:5	32
Spherical Pd (3)	Spherical PbSe (5.8)	Pd: DDT PbSe: OA	0.63	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	Orthorhombic symmetry class	LS	1:3	32
Spherical Ag (4.2)	Spherical PbSe (7.2)	Ag: DDT PbSe: OA	0.66	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	Orthorhombic symmetry class	LS	1:3	32
Spherical PbSe (5.8)	Spherical Bi (14.4)	DDT, OA, or TOPO	-	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	AlB_2-type superlattice	LS_2	-	31
Spherical PbSe (5.8)	Spherical γ-Fe_2O_3 (13.4)	PbSe: OA γ-Fe_2O_3: OA	0.5	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	AlB_2-type superlattice	LS_2	-	32
Spherical $CoPt_3$ (6.2)	Spherical γ-Fe_2O_3 (13.4)	$CoPt_3$: ACA, HDA γ-Fe_2O_3: OA	0.54	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	AlB_2-type superlattice	LS_2	-	32
Spherical Pd (3)	Spherical PbSe (6.7)	Pd: DDT PbSe: OA	0.56	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	AlB_2-type superlattice	LS_2	-	32
Spherical Ag (3.7)	Spherical PbSe (5.8)	Ag: DDT PbSe: OA	0.71	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	AlB_2-type superlattice	LS_2	-	32
Spherical Au (5.5)	Spherical PbSe (7.2)	Au: DDT PbSe: OA	0.787	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	AlB_2-type superlattice	LS_2	-	32
Spherical Au (6.1)	Spherical γ-Fe_2O_3 (11.5)	Au: DDT γ-Fe_2O_3: OA	0.53	Toluene	Solvent evaporation (45 °C)	AlB_2-type superlattice	LS_2		181
Spherical Ag (4.2)	Spherical PbSe (7.2)	DDT, OA, or TOPO	0.66	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	Cu_3Au-type superlattice Fe_4C-type superlattice $NaZn_{13}$-type superlattice Cub-LS_{13}-type superlattice	LS_3 LS_4 LS_{13} LS_{13}	-	31,32
Spherical Ag (3.4)	Spherical PbSe (5.8)	Ag: DDT PbSe: OA	0.68	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	Cu_3Au-type superlattice	LS_3	1:7	32
Spherical Ag (3.6)	Spherical PbSe (6.3)	Ag: DDT PbSe: OA	0.66	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	$CaCu_5$-type superlattice	LS_5	1:7	32
Spherical Ag (3.6)	Spherical PbSe (6.3)	Ag: DDT PbSe: OA	0.66	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	$CaCu_5$-type superlattice	LS_5	1:7	32
Spherical $CoPt_3$ (2.6)	Spherical $CoPt_3$ (4.5)	-	0.71	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation	$CaCu_5$-type superlattice	LS_5	-	406
Spherical Au (5.0)	Spherical PbSe (7.2)	DDT, OA, or TOPO	0.73	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	$CaCu_5$-type superlattice	LS_5	1:7	31
Spherical Au (5)	Spherical γ-Fe_2O_3 (13.4)	Au: DDT γ-Fe_2O_3: OA	0.43	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation (45 °C)	CaB_6-type superlattice	LS_6	1:5	32

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Au (4.6)	Spherical γ-Fe_2O_3 (14.0)	Au: OA γ-Fe_2O_3: DDT	0.4	Trichloroethylene	Solvent evaporation (50 °C)	CaB_6-type superlattice *bcc*-LS_6-type superlattice	LS_6 LS_6	1:8	194
Spherical Au (4.6)	Spherical γ-Fe_2O_3 (14.0)	Au: OA γ-Fe_2O_3: DDT	0.4	Trichloroethylene	Solvent evaporation (50 °C)	(3^2.4.3.4) Archimedean tiling *bcc*-LS_6-type superlattice	LS_4 LS_6	1:5	194
Spherical Au (5.8)	Spherical γ-Fe_2O_3 (14.0)	Au: OA γ-Fe_2O_3: DDT	0.48	Trichloroethylene	Solvent evaporation (50 °C)	*bcc*-LS_6-type superlattice	LS_6	1:8	194
Spherical CdSe (3.4 ± 0.3)	Spherical PbSe (7.3 ± 0.6)	CdSe: TOPO and HDA PbSe: OA	0.56	TCE	Solvent evaporation (70 °C)	AlB_2-type superlattice cub-AB_{13}-type superlattice	LS_2 cub-LS_{13}	-	211
Spherical PbS (3.1)	Spherical CdSe (8)	PbS: OA CdSe: TDPA-HDA-TOPO-TOP	0.47	n-alkanes	Solvent evaporation (85 °C)	NaCl-type superlattice	LS	1:8	41
Spherical PbS (3.1)	Spherical CdSe (8)	PbS: OA CdSe: TDPA-HDA-TOPO-TOP	0.47	n-alkanes	Solvent evaporation (65 °C)	NaCl-type superlattice AlB_2-type superlattice	LS LS_2	1:8	41
Spherical PbS (3.1)	Spherical CdSe (8)	PbS: OA CdSe: TDPA-HDA-TOPO-TOP	0.47	n-alkanes	Solvent evaporation (40 °C)	AlB_2-type superlattice	LS_2	1:8	41
Spherical PbS (3.1)	Spherical CdSe (8)	PbS: OA CdSe: TDPA-HDA-TOPO-TOP	0.47	n-alkanes	Solvent evaporation (25 °C)	AlB_2-type superlattice	LS_2	1:8	41
Spherical PbS (3.1)	Spherical CdSe (8)	PbS: OA CdSe: TDPA-HDA-TOPO-TOP	0.47	n-alkanes	Solvent evaporation (0 °C)	AlB_2-type superlattice	LS_2	1:8	41
Spherical PbS (3.1)	Spherical CdSe (8)	PbS: OA CdSe: TDPA-HDA-TOPO-TOP	0.47	n-alkanes	Solvent evaporation (-20 °C)	$NaZn_{13}$-type superlattice	LS_{13}	1:8	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (100 °C)	CuAu-type superlattice Cu_3Au-type superlattice	LS LS_3	1:9	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (85 °C)	CuAu-type superlattice Cu_3Au-type superlattice Fe_4C-type superlattice	LS LS_3 LS_4	1:9	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (65 °C)	AlB_2-type superlattice Fe_4C-type superlattice	LS_2 LS_4	1:9	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (40 °C)	CuAu-type superlattice AlB_2-type superlattice Cu_3Au-type superlattice	LS LS_2 LS_3	1:9	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (25 °C)	AlB_2-type superlattice	LS_2	1:9	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (0 °C)	AlB_2-type superlattice	LS_2	1:9	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (-20 °C)	CuAu-type superlattice AlB_2-type superlattice Cu_3Au-type superlattice $NaZn_{13}$-type superlattice	LS LS_2 LS_3 LS_{13}	1:9	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (85 °C)	CuAu-type superlattice Cu_3Au-type superlattice	LS LS_3	1:18	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (25 °C)	CuAu-type superlattice AlB_2-type superlattice Cu_3Au-type superlattice Fe_4C-type superlattice $NaZn_{13}$-type superlattice	LS LS_2 LS_3 LS_4 LS_{13}	1:18	41

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (-20 °C)	$NaZn_{13}$-type superlattice	LS_{13}	1:18	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (85 °C)	CuAu-type superlattice Cu_3Au-type superlattice	LS LS_3	1:4.5	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (25 °C)	CuAu-type superlattice AlB_2-type superlattice Cu_3Au-type superlattice	LS LS_2 LS_3	1:4.5	41
Spherical Pd (3.4)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.53	n-alkanes	Solvent evaporation (-20 °C)	NaCl-type superlattice	LS	1:4.5	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (100 °C)	CuAu-type superlattice CsCl-type superlattice	LS LS	1:8	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (85 °C)	CuAu-type superlattice CsCl-type superlattice	LS LS	1:8	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (65 °C)	CuAu-type superlattice $MgZn_2$-type superlattice	LS LS_2	1:8	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (40 °C)	CuAu-type superlattice AlB_2-type superlattice	LS LS_2	1:8	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (25 °C)	$NaZn_{13}$-type superlattice	LS_{13}	1:8	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (85 °C)	CuAu-type superlattice CsCl-type superlattice	LS LS	1:6	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (65 °C)	CuAu-type superlattice AlB_2-type superlattice	LS LS_2	1:6	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (25 °C)	$CaCu_5$-type superlattice	LS_5	1:6	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (85 °C)	CsCl-type superlattice	LS	1:4	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (65 °C)	CuAu-type superlattice AlB_2-type superlattice	LS LS_2	1:4	41
Spherical Pd (4.9)	Spherical PbSe (7.7)	Pd: DDT PbSe: OA	0.74	n-alkanes	Solvent evaporation (25 °C)	CuAu-type superlattice $CaCu_5$-type superlattice	LS LS_5	1:4	41
Spherical CdSe (5.8)	Spherical PbSe (11)	DDT or DDT and OA	0.53	Hexane	Solvent evaporation (70 °C)	AlB_2-type superlattice	LS_2	-	41
Spherical CdSe (5.8)	Spherical PbSe (10.3)	DDT or DDT and OA	0.56	Hexane	Solvent evaporation (70 °C)	AlB_2-type superlattice $NaZn_{13}$-type superlattice	LS_2 LS_{13}	-	41
Spherical CdSe (5.8)	Spherical PbSe (8.0)	DDT or DDT and OA	0.73	Hexane	Solvent evaporation (70 °C)	$MgZn_2$-type superlattice $CaCu_5$-type superlattice	LS_2 LS_5	-	41
Spherical CdSe (5.8)	Spherical PbSe (7.76)	DDT or DDT and OA	0.75	Hexane	Solvent evaporation (70 °C)	$MgZn_2$-type superlattice $CaCu_5$-type superlattice	LS_2 LS_5	-	41
Spherical Au (5.2)	Spherical PbSe (6.98)	DDT or DDT and OA	0.74	Hexane	Solvent evaporation (70 °C)	CsCl-type superlattice $CaCu_5$-type superlattice	LS LS_5	-	41
Spherical Au (5.8)	Spherical PbSe (6.98)	DDT or DDT and OA	0.83	Hexane	Solvent evaporation (70 °C)	CsCl-type superlattice $CaCu_5$-type superlattice	LS LS_5	-	41

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Au (4.1)	Spherical PbS (7)	Au: C_9-length saturated hydrocarbon chains PbS: C_{18}-length saturated hydrocarbon chains	-	Octane	Solvent evaporation (50 °C)	CuAu-type superlattice AlB_2-type superlattice $NaZn_{13}$-type superlattice	LS LS_2 LS_{13}	-	214
Spherical Au (4.1)	Spherical PbS (7)	Au: C_{18}-length saturated hydrocarbon chains PbS: C_9-length saturated hydrocarbon chains	-	Octane	Solvent evaporation (50 °C)	$MgZn_2$-type superlattice $CaCu_5$-type superlattice	LS_2 LS_5	-	214
Spherical Au (4.1)	Spherical γ-Fe_2O_3 (10.2)	Au: C_9-length saturated hydrocarbon chains γ-Fe_2O_3: C_{18}-length saturated hydrocarbon chains	-	Octane	Solvent evaporation (50 °C)	(3^2.4.3.4) Archimedean tiling DDQC *bcc*-LS_6-type superlattice	LS_4 $LS_{3.84}$ LS_6	-	214
Spherical Au (4.1)	Spherical γ-Fe_2O_3 (10.2)	Au: C_{18}-length saturated hydrocarbon chains γ-Fe_2O_3: C_9-length saturated hydrocarbon chains	-	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattices $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	214
Spherical Ag (2.9)	Spherical Ag (11.9)	Ag: OAM	0.42	Toluene	Solvent evaporation (35 °C)	NaCl-type superlattice	LS	1:2 1:4 1:10	217
Spherical Ag (3.7)	Spherical Ag (11.9)	Ag: OAM	0.44	Toluene	Solvent evaporation (35 °C)	NaCl-type superlattice	LS	1:2 1:4 1:10	217
Spherical Ag (3.7)	Spherical Ag (9.6)	Ag: OAM	0.52	Toluene	Solvent evaporation (35 °C)	AlB_2-type superlattice	LS_2	1:2 1:4 1:10	217
Spherical Ag (2.9)	Spherical Ag (8.2)	Ag: OAM	0.55	Toluene	Solvent evaporation (35 °C)	AlB_2-type superlattice	LS_2	1:2 1:4 1:10	217
Spherical Ag (3.7)	Spherical Ag (8.2)	Ag: OAM	0.58	Toluene	Solvent evaporation (35 °C)	AlB_2-type superlattice	LS_2	1:2	217
Spherical Ag (3.7)	Spherical Ag (8.2)	Ag: OAM	0.58	Toluene	Solvent evaporation (35 °C)	AlB_2-type superlattice $NaZn_{13}$-type superlattice	LS_2 LS_{13}	1:4	217
Spherical Ag (3.7)	Spherical Ag (8.2)	Ag: OAM	0.58	Toluene	Solvent evaporation (35 °C)	$NaZn_{13}$-type superlattice	LS_{13}	1:10	217
Spherical Ag (3.7)	Spherical Ag (7.5)	Ag: OAM	0.64	Toluene	Solvent evaporation (35 °C)	$NaZn_{13}$-type superlattice	LS_{13}	1:2 1:4 1:10	217
Spherical Ag (3.7)	Spherical Ag (5.5)	Ag: OAM	0.79	Toluene	Solvent evaporation (35 °C)	$MgZn_2$-type superlattice	LS_2	1:2 1:4	217
Spherical Ag (5.5)	Spherical Ag (7.5)	Ag: OAM	0.81	Toluene	Solvent evaporation (35 °C)	$MgZn_2$-type superlattice	LS_2	1:2 1:4 1:10	217

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Ag (4.0)	Spherical Ag (11.9)	Small Ag particles: DDT Large Ag particles: OAM	0.42	Toluene	Solvent evaporation (35 °C)	CaB_6-type superlattice DDQC	LS_6 $LS_{3.84}$	1:2 1:4	217
Spherical Ag (4.0)	Spherical Ag (11.9)	Small Ag particles: DDT Large Ag particles: OAM	0.42	Toluene	Solvent evaporation (35 °C)	CaB_6-type superlattice	LS_6	1:10	217
Spherical Au (4.0)	Spherical Ag (11.9)	Small Au particles: DDT Large Ag particles: OAM	0.42	Toluene	Solvent evaporation (35 °C)	CaB_6-type superlattice DDQC	LS_6 $LS_{3.84}$	1:4	217
Spherical Ag (3.7)	Spherical $CoFe_2O_4$ (12)	Ag: OAM $CoFe_2O_4$: OA	0.42	Toluene	Solvent evaporation (35 °C)	AlB_2-type superlattice (3^2.4.3.4) Archimedean tiling DDQC	LS_2 LS_4 $LS_{3.84}$	1:4	217
Spherical Ag (4)	Spherical $CoFe_2O_4$ (12)	Ag: DDT $CoFe_2O_4$: OA	0.42	Toluene	Solvent evaporation (35 °C)	CaB_6-type superlattice DDQC	LS_6 $LS_{3.84}$	1:4	217
Spherical Au (3.1)	Spherical PbS (6.3)	Au: nonanethiol (λ=0.77) PbS: nonanoic acid (λ=0.38)	0.57	Octane	Solvent evaporation (50 °C)	$NaZn_{13}$-type superlattice	LS_{13}	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: DDT (λ=1.01) PbS: nonanoic acid (λ=0.38)	0.61	Octane	Solvent evaporation (50 °C)	$NaZn_{13}$-type superlattice	LS_{13}	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: nonanethiol (λ=0.77) PbS: myristic acid (λ=0.57)	0.53	Octane	Solvent evaporation (50 °C)	AlB_2-type superlattice	LS_2	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: DDT (λ=1.01) PbS: myristic acid (λ=0.57)	0.56	Octane	Solvent evaporation (50 °C)	AlB_2-type superlattice	LS_2	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: Octadecanethiol (λ=1.47) PbS: myristic acid (λ=0.57)	0.62	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: hexanethiol (λ=0.54) PbS: OA (λ=0.66)	0.47	Octane	Solvent evaporation (50 °C)	NaCl-type superlattice	LS	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: nonanethiol (λ=0.77) PbS: OA (λ=0.66)	0.51	Octane	Solvent evaporation (50 °C)	CuAu-type superlattice	LS	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: DDT (λ=1.01) PbS: OA (λ=0.66)	0.54	Octane	Solvent evaporation (50 °C)	Cu_3Au-type superlattice $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	221
Spherical Au (3.1)	Spherical PbS (6.3)	Au: Octadecanethiol (λ=1.47) PbS: OA (λ=0.66)	0.60	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (4.3)	Spherical PbS (6.3)	Au: DDT (λ=0.73) PbS: nonanoic acid (λ=0.38)	0.78	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (4.3)	Spherical PbS (6.3)	Au: Octadecanethiol (λ=1.06) PbS: myristic acid (λ=0.57)	0.79	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (5.1)	Spherical PbS (6.3)	Au: pentadecanethiol (λ=0.75) PbS: myristic acid (λ=0.57)	0.86	Octane	Solvent evaporation (50 °C)	$MgZn_2$-type superlattice $CaCu_5$-type superlattice	LS_2 LS_5	-	221
Spherical Au (5.1)	Spherical PbS (6.3)	Au: DDT (λ=0.61) PbS: OA (λ=0.66)	0.80	Octane	Solvent evaporation (50 °C)	$MgZn_2$-type superlattice $CaCu_5$-type superlattice	LS_2 LS_5	-	221
Spherical Au (3.1)	Spherical PbS (7.5)	Au: hexanethiol (λ=0.54) PbS: OA (λ=0.55)	0.41	Octane	Solvent evaporation (50 °C)	NaCl-type superlattice	LS	-	221
Spherical Au (3.1)	Spherical PbS (7.5)	Au: nonanethiol (λ=0.77) PbS: OA (λ=0.55)	0.44	Octane	Solvent evaporation (50 °C)	CuAu-type superlattice	LS	-	221
Spherical Au (3.1)	Spherical PbS (7.5)	Au: DDT (λ=1.01) PbS: OA (λ=0.55)	0.47	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattice	LS_3	-	221
Spherical Au (3.1)	Spherical PbS (7.5)	Au: Octadecanethiol (λ=1.47) PbS: OA (λ=0.55)	0.52	Octane	Solvent evaporation (50 °C)	$NaZn_{13}$-type superlattice *bcc*-LS_6-type superlattice	LS_{13} LS_6	-	221

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Au (4.3)	Spherical PbS (7.5)	Au: nonanethiol (λ=0.56) PbS: OA (λ=0.55)	0.57	Octane	Solvent evaporation (50 °C)	CuAu-type superlattice AlB_2-type superlattice	LS LS_2	-	221
Spherical Au (4.3)	Spherical PbS (7.5)	Au: DDT (λ=0.73) PbS: OA (λ=0.55)	0.61	Octane	Solvent evaporation (50 °C)	CuAu-type superlattice $NaZn_{13}$-type superlattice	LS LS_{13}	-	221
Spherical Au (4.3)	Spherical PbS (7.5)	Au: pentadecanethiol (λ=0.89) PbS: OA (λ=0.55)	0.64	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (4.3)	Spherical PbS (7.5)	Au: Octadecanethiol (λ=1.06) PbS: OA (λ=0.55)	0.67	Octane	Solvent evaporation (50 °C)	$MgZn_2$-type superlattice $CaCu_5$-type superlattice	LS_2 LS_5	-	221
Spherical Au (5.1)	Spherical PbS (7.5)	Au: DDT (λ=0.61) PbS: OA (λ=0.55)	0.69	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (5.1)	Spherical PbS (7.5)	Au: pentadecanethiol (λ=0.75) PbS: OA (λ=0.55)	0.73	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (5.1)	Spherical PbS (7.5)	Au: Octadecanethiol (λ=0.89) PbS: OA (λ=0.55)	0.76	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: hexanethiol (λ=0.54) PbS: Undecanoic acid (λ=0.31)	0.37	Octane	Solvent evaporation (50 °C)	NaCl-type superlattice	LS	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: nonanethiol (λ=0.77) PbS: Undecanoic acid (λ=0.31)	0.40	Octane	Solvent evaporation (50 °C)	CaB_6-type superlattice	LS_6	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: DDT (λ=1.01) PbS: Undecanoic acid (λ=0.31)	0.43	Octane	Solvent evaporation (50 °C)	*bcc*-LS_6-type superlattice	LS_6	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: Octadecanethiol (λ=1.47) PbS: Undecanoic acid (λ=0.31)	0.47	Octane	Solvent evaporation (50 °C)	*bcc*-LS_6-type superlattice	LS_6	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: hexanethiol (λ=0.54) PbS: OA (λ=0.45)	0.35	Octane	Solvent evaporation (50 °C)	NaCl-type superlattice	LS	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: nonanethiol (λ=0.77) PbS: OA (λ=0.45)	0.37	Octane	Solvent evaporation (50 °C)	CaB_6-type superlattice	LS_6	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: DDT (λ=1.01) PbS: OA (λ=0.45)	0.40	Octane	Solvent evaporation (50 °C)	*bcc*-LS_6-type superlattice	LS_6	-	221
Spherical Au (3.1)	Spherical PbS (9.3)	Au: Octadecanethiol (λ=1.47) PbS: OA (λ=0.45)	0.44	Octane	Solvent evaporation (50 °C)	*bcc*-LS_6-type superlattice	LS_6	-	221
Spherical Au (4.3)	Spherical PbS (9.3)	Au: nonanethiol (λ=0.56) PbS: Undecanoic acid (λ=0.31)	0.52	Octane	Solvent evaporation (50 °C)	CaCu-type superlattice DDQC	LS $LS_{3.84}$	-	221
Spherical Au (4.3)	Spherical PbS (9.3)	Au: DDT (λ=0.73) PbS: Undecanoic acid (λ=0.31)	0.55	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattice $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	221
Spherical Au (4.3)	Spherical PbS (9.3)	Au: pentadecanethiol (λ=0.89) PbS: Undecanoic acid (λ=0.31)	0.57	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattice $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	221
Spherical Au (4.3)	Spherical PbS (9.3)	Au: Octadecanethiol (λ=1.06) PbS: Undecanoic acid (λ=0.31)	0.60	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattice $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	221
Spherical Au (4.3)	Spherical PbS (9.3)	Au: nonanethiol (λ=0.56) PbS: OA (λ=0.45)	0.48	Octane	Solvent evaporation (50 °C)	CuAu-type superlattice DDQC	LS $LS_{3.84}$	-	221
Spherical Au (4.3)	Spherical PbS (9.3)	Au: DDT (λ=0.73) PbS: OA (λ=0.45)	0.51	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattice $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	221
Spherical Au (4.3)	Spherical PbS (9.3)	Au: pentadecanethiol (λ=0.89) PbS: OA (λ=0.45)	0.54	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattice $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	221

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Au (4.3)	Spherical PbS (9.3)	Au: Octadecanethiol (λ=1.06) PbS: OA (λ=0.45)	0.56	Octane	Solvent evaporation (50 °C)	Li_3Bi-type superlattice $NaZn_{13}$-type superlattice	LS_3 LS_{13}	-	221
Spherical Au (5.1)	Spherical PbS (9.3)	Au: nonanethiol (λ=0.47) PbS: Undecanoic acid (λ=0.31)	0.59	Octane	Solvent evaporation (50 °C)	AlB_2-type superlattice $NaZn_{13}$-type superlattice	LS_2 LS_{13}	-	221
Spherical Au (5.1)	Spherical PbS (9.3)	Au: nonanethiol (λ=0.47) PbS: OA (λ=0.45)	0.55	Octane	Solvent evaporation (50 °C)	AlB_2-type superlattice	LS_2	-	221
Spherical Au (5.1)	Spherical PbS (9.3)	Au: DDT (λ=0.61) PbS: OA (λ=0.45)	0.58	Octane	Solvent evaporation (50 °C)	CuAu-type superlattice Cu_3Au-type superlattice	LS LS_3	-	221
Spherical Au (5.1)	Spherical PbS (9.3)	Au: Octadecanethiol (λ=0.89) PbS: OA (λ=0.45)	0.64	Octane	Solvent evaporation (50 °C)	$CaCu_5$-type superlattice	LS_5	-	221
Spherical Au (5)	Spherical γ-Fe_2O_3 (13.4)	Au: DDT Fe_3O_4: OA	0.43 ± 0.01	TCE	Solvent evaporation (50 °C)	AlB_2-type superlattice DDQC (3^2.4.3.4) Archimedean tiling (3^3.4^2) Archimedean tiling CaB_6-type superlattice	LS_2 $LS_{3.84}$ LS_4 LS_4 LS_6	-	33
Spherical Au (4.7)	Spherical γ-Fe_2O_3 (12.6)	Au: DDT Fe_3O_4: OA	0.43 ± 0.01	TCE	Solvent evaporation (50 °C)	DDQC	$LS_{3.84}$	-	33
Spherical Pd (3)	Spherical PbS (9)	-	0.43 ± 0.01	TCE	Solvent evaporation (50 °C)	DDQC	$LS_{3.84}$	-	33
Spherical {Mo_{132}} (2.9)	Spherical PbS (10.7)	Mo_{132}: DDA PbS: OA	0.41	Toluene and chlorobenzene	Solvent evaporation (50 °C)	NaCl-type superlattice AlB_2-type superlattice *bcc*-LS_6-type superlattice $NaZn_{13}$-type superlattice	LS LS_2 LS_6 LS_{13}	-	206
Spherical {Mo_{132}} (2.9)	Spherical PbS (8.1)	Mo_{132}: DDA PbS: OA	0.51	Toluene and chlorobenzene	Solvent evaporation (50 °C)	NaCl-type superlattice AlB_2-type superlattice DDQC $NaZn_{13}$-type superlattice	LS LS_2 $LS_{3.84}$ LS_{13}	-	206
Spherical {Mo_{132}} (2.9)	Spherical PbS (6.6)	Mo_{132}: DDA PbS: OA	0.58	Toluene and chlorobenzene	Solvent evaporation (50 °C)	NaCl-type superlattice AlB_2-type superlattice $NaZn_{13}$-type superlattice	LS LS_2 LS_{13}	-	206
Spherical {Mo_{132}} (2.9)	Spherical PbS (4.7)	Mo_{132}: DDA PbS: OA	0.74	Toluene and chlorobenzene	Solvent evaporation (50 °C)	NaCl-type superlattice AlB_2-type superlattice $CaCu_5$-type superlattice $NaZn_{13}$-type superlattice	LS LS_2 LS_5 LS_{13}	-	206
Spherical Ag (4.0)	Spherical amorphous-phase Co (9.2)	Ag: DDT Co: OA	0.49	Toluene	Solvent evaporation (25 °C)	NaCl-type superlattice	LS	2:1	208
Spherical Ag (4.0)	Spherical *hcp*-phase Co (9.3)	Ag: DDT Co: OA	0.49	Toluene	Solvent evaporation (25 °C)	CuAu-type superlattice Cu_3Au-type superlattice DDQC $NaZn_{13}$-type superlattice	LS LS_3 $LS_{3.84}$ LS_{13}	2:1	208
Spherical Ag (4.0)	Spherical amorphous-phase Co (9.2)	Ag: DDT Co: OA	0.49	Toluene	Solvent evaporation (65 °C)	AlB_2-type superlattice Cu_3Au-type superlattice	LS_2 LS_3	2:1	208
Spherical Ag (4.0)	Spherical *hcp*-phase Co (9.3)	Ag: DDT Co: OA	0.49	Toluene	Solvent evaporation (65 °C)	AlB_2-type superlattice Cu_3Au-type superlattice	LS_2 LS_3	2:1	208

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Ag (4.0)	Spherical amorphous-phase Co (7.2)	Ag: DDT Co: OA	0.59	Toluene	Solvent evaporation (25 °C)	AlB_2-type superlattice	LS_2	2:1	208
Spherical Ag (4.0)	Spherical *hcp*-phase Co (7.1)	Ag: DDT Co: OA	0.59	Toluene	Solvent evaporation (25 °C)	AlB_2-type superlattice	LS_2	2:1	208
Spherical Au (4.5)	Spherical hollow core-shell iron/iron oxide (11.7)	-	-	Toluene and TCE	Solvent evaporation (40-50 °C)	NaCl-type superlattice	LS	1:2	241
Spherical Au (4.5)	Spherical hollow core-shell iron/iron oxide (11.7)	DDT	-	Toluene and TCE	Solvent evaporation (40-50 °C)	NiAs-type superlattice	LS	1:2	241
Spherical Au (4.5)	Spherical hollow core-shell iron/iron oxide (11.7)	DDT	-	Toluene and TCE	Solvent evaporation (40-50 °C)	NiAs-type superlattice AlB_2-type superlattice	LS LS_2	1:2	241
Spherical Au (4.5)	Spherical hollow core-shell iron/iron oxide (11.7)	-	-	Toluene and TCE	Solvent evaporation (40-50 °C)	AlB_2-type superlattice	LS_2	1:4-5	241
Spherical Au (4.5)	Spherical hollow core-shell iron/iron oxide (11.7)	OA	-	Toluene and TCE	Solvent evaporation (40-50 °C)	AlB_2-type superlattice	LS_2	1:3	241
Spherical Au (4.5)	Spherical hollow core-shell iron/iron oxide (11.7)	-	-	Toluene and TCE	Solvent evaporation (40-50 °C)	$NaZn_{13}$-type superlattice cub-AB_{13}-type superlattice	LS_{13} LS_{13}	1:6-8	241
Spherical Au (4.5)	Spherical hollow core-shell iron/iron oxide (11.7)	OAM	-	Toluene and TCE	Solvent evaporation (40-50 °C)	$NaZn_{13}$-type superlattice cub-AB_{13}-type superlattice	LS_{13} LS_{13}	1:6-8	241
Spherical Ag (4.0)	Spherical Ag (11.9)	Small Ag particles: DDT Large Ag particles: OAM	0.42	Hexane (on DEG)	Air-liquid interface	NaCl-type superlattice	LS	1:4	217
Spherical FePt (6)	Spherical Fe_3O_4 (15)	-	-	Hexane (on DEG)	Air-liquid interface	AlB_2-type superlattice Cu_3Au-type superlattice $NaZn_{13}$-type superlattice	LS_2 LS_3 LS_{13}	1:3	248
Spherical Au (3.8)	Spherical Fe_3O_4 (13.4)	Au: Atactic PS (M_n = 3.0 k) Fe_3O_4: Atactic PS (M_n = 5.3 k)	0.49	Toluene (on DEG)	Air-liquid interface	$NaZn_{13}$-type superlattice	LS_{13}	1:15	254
Spherical Au (3.8)	Spherical Fe_3O_4 (13.4)	Au: Atactic PS (M_n = 3.0 k) Fe_3O_4: Atactic PS (M_n = 5.3 k)	0.49	Toluene (on DEG)	Air-liquid interface	*bcc*-LS_6-type superlattice	LS_6	1:7	254
Spherical Au (4.2)	Spherical Fe_3O_4 (13.4)	Au: Atactic PS (M_n = 1.1 k) Fe_3O_4: Atactic PS (M_n = 5.3 k)	0.40	Toluene (on DEG)	Air-liquid interface	NaCl-type superlattice	LS	-	254
Spherical Au (4.2)	Spherical Fe_3O_4 (13.4)	Au: Atactic PS (M_n = 5.3 k) Fe_3O_4: Atactic PS (M_n = 5.3 k)	0.69	Toluene (on DEG)	Air-liquid interface	$MgZn_2$-type superlattice $CaCu_5$-type superlattice	LS_2 LS_5	-	254
Spherical Au (6.4)	Spherical Fe_3O_4 (16.5)	OA and/or OAM	0.44	Hexane (on DEG)	Air-liquid interface	2D LS-type superlattice	LS	-	253

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical FePt (5.5)	Spherical Fe_3O_4 (16.5)	OA and/or OAM	0.40	Hexane (on DEG)	Air-liquid interface	2D LS-type superlattice	LS	-	253
Spherical Fe_3O_4 (13.5)	Spherical $NaYF_4$:Yb/Er (28.9)	OA and/or OAM	0.49	Hexane (on DEG)	Air-liquid interface	2D LS-type superlattice	LS	-	253
Spherical FePt (3.1)	Spherical Fe_3O_4 (16.5)	OA and/or OAM	0.26	Hexane (on DEG)	Air-liquid interface	2D LS_2-type superlattice	LS_2	-	253
Spherical Au (6.4)	Spherical Fe_3O_4 (11.2)	OA and/or OAM	0.63	Hexane (on DEG)	Air-liquid interface	2D L_2S_3-type superlattice	L_2S_3	-	253
Spherical FePt (6.0)	Spherical Fe_3O_4 (16.5)	OA and/or OAM	0.42	Hexane (on DEG)	Air-liquid interface	2D LS_3-type superlattice	LS_3	-	253
Spherical Ag (4.8)	Spherical Au (5.1)	Ag: TMA Au: MUA	0.97	Water	Electrostatic	Diamond-type (or sphalerite-type) superlattice	LS	-	49
Spherical Au (11.4)	Spherical Au (11.4)	DNA	-	Water	DNA-guided	CsCl-type superlattice	LS	-	278
Spherical Au (12.5)	Spherical Au (12.5)	DNA	-	Water	DNA-guided	CsCl-type superlattice	LS	-	278
Spherical Au (5)	Spherical Au (9.7)	DNA (DNA linker ratio = 3)	0.75	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (2.5)	Spherical Au (4.4)	DNA (DNA linker ratio = 0.4)	0.61	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (5)	Spherical Au (9.5)	DNA (DNA linker ratio = 0.5)	0.76	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (5)	Spherical Au (10.2)	DNA (DNA linker ratio = 0.5)	1	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (5)	Spherical Au (10.2)	DNA (DNA linker ratio = 0.5)	0.85	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (5)	Spherical Au (15.8)	DNA (DNA linker ratio = 0.2)	0.99	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (9.7)	Spherical Au (19.2)	DNA (DNA linker ratio = 0.3)	0.98	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (14.8)	Spherical Au (24.5)	DNA (DNA linker ratio = 0.3)	0.99	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (14.8)	Spherical Au (29)	DNA (DNA linker ratio = 0.2)	0.94	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (19.2)	Spherical Au (29)	DNA (DNA linker ratio = 0.4)	0.99	water	DNA-guided	CsCl-type superlattice	LS	-	256
Spherical Au (2.5)	Spherical Au (4.4)	DNA (DNA linker ratio = 2)	0.6	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (4.6)	Spherical Au (4.6)	DNA (DNA linker ratio = 1)	0.56	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (4.9)	Spherical Au (4.9)	DNA (DNA linker ratio = 1)	0.38	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (4.9)	Spherical Au (9.5)	DNA (DNA linker ratio = 1.5)	0.55	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical Au (4.9)	Spherical Au (9.5)	DNA (DNA linker ratio = 1.5)	0.47	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (5)	Spherical Au (9.7)	DNA (DNA linker ratio = 3)	0.51	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (4.9)	Spherical Au (9.5)	DNA (DNA linker ratio = 1.5)	0.41	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (9.8)	Spherical Au (14.4)	DNA (DNA linker ratio = 3)	0.55	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (9.7)	Spherical Au (15.8)	DNA (DNA linker ratio = 2.4)	0.64	water	DNA-guided	AlB_2-type superlattice	LS_2	-	256
Spherical Au (4.6)	Spherical Au (7.5)	DNA (DNA linker ratio = 2)	0.37	water	DNA-guided	Cr_3Si-type superlattice	LS_3	-	256
Spherical Au (4.6)	Spherical Au (7.5)	DNA (DNA linker ratio = 2.5)	0.51	water	DNA-guided	Cr_3Si-type superlattice	LS_3	-	256
Spherical Au (9.8)	Spherical Au (14.4)	DNA (DNA linker ratio = 2.25)	0.43	water	DNA-guided	Cr_3Si-type superlattice	LS_3	-	256
Spherical Au (4.6)	Spherical Au (9.7)	DNA (DNA linker ratio = 3)	0.35	water	DNA-guided	Cr_3Si-type superlattice	LS_3	-	256
Spherical Au (4.6)	Spherical Au (9.8)	DNA (DNA linker ratio = 3)	0.48	water	DNA-guided	*bcc*-LS_6-type superlattice	LS_6	-	256
Spherical Au (7.5)	Spherical Au (15.8)	DNA (DNA linker ratio = 2.25)	0.37	water	DNA-guided	*bcc*-LS_6-type superlattice	LS_6	-	256
Spherical Fe_3O_4 (5.4)	Spherical Fe_3O_4 (13.5)	5.4 nm Fe_3O_4: Pentaethylenehexamine-terminated polystyrene 13.5 nm Fe_3O_4: Diethylenetriamine-terminated polystyrene	0.61	Toluene	Spin-coating	$NaZn_{13}$-type superlattice	LS_{13}	-	308
Spherical Au (4)	Spherical Au (8)	1-hexadecanethiol	-	Hexane	Emulsion-assisted (Hexane-in-water, surfactant: Triton X-100)	$NaZn_{13}$-type superlattice	LS_{13}	-	250
Spherical Au (5)	Spherical γ-Fe_2O_3 (10)	OA or DDT	0.5	Hexane	Emulsion-assisted (Hexane-in-water)	$NaZn_{13}$-type superlattice	LS_{13}	1:15	315
Spherical $CoFe_2O_4$ (8.0)	Spherical Fe_3O_4 (17)	-	-	Hexane	Emulsion-assisted (Hexane-in-water, surfactant: DTAB)	$NaZn_{13}$-type superlattices	LS_{13}	1:13	319
Spherical $CoFe_2O_4$ (8.5)	Spherical Fe_3O_4 (17)	-	-	Hexane	Emulsion-assisted (Hexane-in-water, surfactant: DTAB)	$NaZn_{13}$-type superlattices	LS_{13}	1:13	319
Spherical $CoFe_2O_4$ (4.5)	Spherical Fe_3O_4 (11)	-	-	Hexane	Emulsion-assisted (Hexane-in-water, surfactant: DTAB)	AlB_2-type superlattices	LS_2	-	319
Spherical Fe_3O_4 (6.5)	Spherical Fe_3O_4 (9)	-	-	Hexane	Emulsion-assisted (Hexane-in-water, surfactant: DTAB)	$MgZn_2$-type superlattices	LS_2	-	319

Table S1. Binary superstructures formed through the co-assembly of spherical nanoparticles (continuation).

Spherical $CoFe_2O_4$ (4.5)	Spherical Fe_3O_4 (7)	-	-	Hexane	Emulsion-assisted (Hexane-in-water, surfactant: DTAB)	$CaCu_5$-type superlattices	LS_5	-	319
Spherical $CoFe_2O_4$ (4.5)	Spherical Fe_3O_4 (15)	-	-	Hexane	Emulsion-assisted (Hexane-in-water, surfactant: DTAB)	NaCl-type superlattices	LS	-	319
Spherical CdSe (8)	Spherical $CoFe_2O_4$ (20)	OA	0.48	TCE	Emulsion-assisted (TCE-in-FC-40, or TCE-in-FC-70, microfluidic)	AlB_2-type superlattices	LS_2	-	149
Spherical PbS (10)	Spherical $CoFe_2O_4$ (20)	OA	0.54	TCE	Emulsion-assisted (TCE-in-FC-40, or TCE-in-FC-70, microfluidic)	AlB_2-type superlattices	LS_2	-	149

Notation: (ϰ) The size refers to that of the core particles; (†) The size ratio (γ) is the effective size ratio, calculated using the effective diameters (d_{eff}, where $d_{eff} = d_{core} + 2d_{shell}$, being d_{core} the core diameter and d_{shell} the shell thickness); DDT, dodecanethiol; OA, oleic acid; TOPO, tri-n-octylphosphine oxide; ODA, octadecylamine; ODPA, octadecylphosphonic acid; TCE, tetrachloroethylene; RT, room temperature; ACA, 1-adamantanecarboxylic acid; HDA, hexadecylamine; TOP, trioctylphosphine; TDPA, tetradecylphosphonic acid; DDQC, dodecagonal quasicrystal; OAM, oleylamine; C_{10}-thiol, alkanethiol; DEG, diethylene glycol; TMA, $HS(CH_2)_{11}NMe_3^+Cl^-$; MUA, $HS(CH_2)_{10}COOH$; FC-40, perfluoro-di-n-butylmethylamine; FC-70, perfluorotripentylamine; DDA, dodecyldimethylammonium cations; DEG, diethylene glycol; DDQC, dodecagonal quasicrystal; DTAB, dodecyltrimethylammonium bromide; DNA, deoxyribonucleic acid.

Table 2. Binary superstructures formed through the co-assembly of anisotropic nanoparticles.

Small particles (S) (size in nm)$^{(x)}$	Large particles (L) (size in nm)$^{(x)}$	Particle functionalization	Size ratio (γ)$^{(f)}$	Solvent	Co-assembly approach	Superlattice	Stoichiometry	n_L:n_S	Ref.
Truncated cuboctahedra Au (4.5) Resembling spherical shape	Icosahedral or decahedral Au (7.8) Resembling spherical shape	C_{10}-thiol	0.58	Toluene	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	24
Spherical Au (3)	LaF_3 triangular nanoplates (9)	DDT, OA, or TOPO	0.33	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation	-	-	-	31
Spherical Au (3)	LaF_3 triangular nanoplates (9)	DDT, OA, or TOPO	0.33	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation	-	-	-	31
Spherical Au (6.2)	LaF_3 triangular nanoplates (9)	DDT, OA, or TOPO	0.69	Toluene or toluene mixture with TCE or chloroform	Solvent evaporation	-	-	-	31
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (12.5)	$CsPbBr_3$: DDAB LaF_3: OA	0.430	Toluene or octane	Solvent evaporation (RT)	Columnar LS(I)-type superlattice: vertically oriented stacks of nanodisks created a simple square columnar lattice with *p4mm* plane group symmetry, while nanocube pillars filled the space between the disk columns	LS	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (13.3)	$CsPbBr_3$: DDAB LaF_3: OA	0.408	Toluene or octane	Solvent evaporation (RT)	Columnar LS(I)-type superlattice	LS	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (16.6)	$CsPbBr_3$: DDAB LaF_3: OA	0.337	Toluene or octane	Solvent evaporation (RT)	Columnar LS(I)-type superlattice	LS	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (18.5)	$CsPbBr_3$: DDAB LaF_3: OA	0.306	Toluene or octane	Solvent evaporation (RT)	Columnar LS(I)-type superlattice	LS	-	234
Cubic $CsPbBr_3$ (8.6)	LaF_3 nanodisks (26.5)	$CsPbBr_3$: DDAB LaF_3: OA	0.335	Toluene or octane	Solvent evaporation (RT)	Columnar LS(I)-type superlattice	LS	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (26.5)	$CsPbBr_3$: DDAB LaF_3: OA	0.221	Toluene or octane	Solvent evaporation (RT)	Columnar LS_2(I)-type superlattice with a *c2mm* symmetry: two nanocube pillars filled the spaces between every four nanodisk columns	LS_2	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (12.5)	$CsPbBr_3$: DDAB LaF_3: OA	0.430	Toluene or octane	Solvent evaporation (RT)	Columnar LS_2(II)-type superlattice: a centered rectangular lattice with LS_2 stoichiometry and *c2mm* symmetry	LS_2	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (12.5)	$CsPbBr_3$: DDAB LaF_3: OA	0.430	Toluene or octane	Solvent evaporation (RT)	Columnar LS_4-type superlattice: columns of nanodisks arranged in a simple hexagonal lattice were uniformly surrounded by nanocubes	LS_4	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (21.0)	$CsPbBr_3$: DDAB LaF_3: OA	0.273	Toluene or octane	Solvent evaporation (RT)	Columnar LS_6-type superlattice: 12 distinct pillars of nanocubes arranged in a dodecagonal pattern around columns of nanodisks	LS_6	-	234

Table 2. Binary superstructures formed through the co-assembly of anisotropic nanoparticles (continuation).

Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (28.4)	$CsPbBr_3$: DDAB LaF_3: OA	0.207	Toluene or octane	Solvent evaporation (RT)	Columnar LS_6-type superlattice	LS_6	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (9.2)	$CsPbBr_3$: DDAB LaF_3: OA	0.555	Toluene or octane	Solvent evaporation (RT)	Columnar LS(II)-type superlattice: pairs of nanocube columns occupied interstitial sites created by six columns of adjacent disks	LS	-	234
Cubic $CsPbBr_3$ (8.6)	LaF_3 nanodisks (18.5)	$CsPbBr_3$: DDAB LaF_3: OA	0.463	Toluene or octane	Solvent evaporation (RT)	Lamellar LS(I)-type superlattice: alternated one-dimensional strings of nanodisks and nanocubes stacked face-to-face on the substrate	LS	-	234
Cubic $CsPbBr_3$ (5.3)	LaF_3 nanodisks (6.5)	$CsPbBr_3$: DDAB LaF_3: OA	-	Toluene or octane	Solvent evaporation (RT)	Lamellar L_2S-type superlattice: each chain of nanocubes was surrounded by six chains of nanodisks	L_2S	-	234
Cubic $CsPbBr_3$ (8.6)	LaF_3 nanodisks (12.5)	$CsPbBr_3$: DDAB LaF_3: OA	-	Toluene or octane	Solvent evaporation (RT)	NaCl-type superlattice: clusters of three disks occupying individual lattice sites in the *fcc* sublattice	LS	-	234
Cubic $CsPbBr_3$ (8.6)	LaF_3 nanodisks (9)	$CsPbBr_3$: DDAB LaF_3: OA	-	Toluene or octane	Solvent evaporation (RT)	ReO_3-type superlattice: the nanocubes occupy the primitive positions of the unit cell, forming a simple cubic arrangement, while the nanodisks are positioned between the nanocubes, aligned face-to-face	LS_3	-	234
Cubic $CsPbBr_3$ (8.6)	LaF_3 nanodisks (6.5)	$CsPbBr_3$: DDAB LaF_3: OA	-	Toluene or octane	Solvent evaporation (RT)	Disrupted ReO_3-type superlattice	LS_3	-	234
Cubic $CsPbBr_3$ (8.6)	Spherical $NaGdF_4$ (18.6)	$CsPbBr_3$: DDAB $NaGdF_4$: OA	0.439	Toluene	Solvent evaporation (RT)	NaCl-type superlattice	LS	1:1.2	240
Cubic $CsPbBr_3$ (8.6)	Spherical $NaGdF_4$ (16.5)	$CsPbBr_3$: DDAB $NaGdF_4$: OA	0.495	Toluene	Solvent evaporation (RT)	$CaTiO_3$-type superlattice	LS_4	1:4.2	240
Cubic $CsPbBr_3$ (8.6)	Spherical $NaGdF_4$ (16.5)	$CsPbBr_3$: DDAB $NaGdF_4$: OA	0.495	Toluene	Solvent evaporation (RT)	AlB_2-type superlattice	LS_2	1:2.2	240
Cubic $CsPbBr_3$ (8.6)	Spherical Fe_3O_4 (19.8)	$CsPbBr_3$: DDAB Fe_3O_4: OA	0.414	Toluene	Solvent evaporation (RT)	AlB_2-type superlattice	LS_2	1:2.2	240
Cubic $CsPbBr_3$ (5.3)	Spherical Fe_3O_4 (11.2-15.6)	$CsPbBr_3$: DDAB Fe_3O_4: OA	0.443 - 0.336	Toluene	Solvent evaporation (RT)	AlB_2-type superlattice AB_2-type superlattice	LS_2 LS_2	1:2.2	240
Cubic $CsPbBr_3$ (5.3)	Spherical Fe_3O_4 (16.9)	$CsPbBr_3$: DDAB Fe_3O_4: OA	0.315	Toluene	Solvent evaporation (RT)	ABO_6-type superlattice	LS_7	1:12	240
Cubic $CsPbBr_3$ (5.3)	Spherical Fe_3O_4 (15.2)	$CsPbBr_3$: DDAB Fe_3O_4: OA	0.344	Toluene	Solvent evaporation (RT)	NaCl-type superlattice AlB_2-type superlattice AB_2-type superlattice $CaTiO_3$-type superlattice	LS LS_2 LS_2 LS_4	-	240
Cubic $CsPbBr_3$ (8.6)	Truncated cuboidal PbS (10.7-11.7)	$CsPbBr_3$: DDAB PbS: OA	0.72–0.78	Toluene	Solvent evaporation (RT)	NaCl-type superlattice CuAu-type superlattice $CaTiO_3$-type superlattice	LS LS LS_4	-	240

Table 2. Binary superstructures formed through the co-assembly of anisotropic nanoparticles (continuation).

Cubic $CsPbBr_3$ (5.3)	Truncated cuboidal PbS (10.7)	$CsPbBr_3$: DDAB PbS: OA	-	Toluene	Solvent evaporation (RT)	AlB_2-type superlattice	LS_2	-	240
Cubic $CsPbBr_3$ (8.6)	Disk-shaped $NaGdF_4$ (31.5 nm in diameter and 18.5 nm thick)	$CsPbBr_3$: DDAB $NaGdF_4$: OA	-	Toluene	Solvent evaporation (RT)	CaC_2-type superlattice	LS_2	-	240
Cubic $FAPbBr_3$ (9)	Spherical $NaGdF_4$ (15.1-19.5)	$FAPbBr_3$: - $NaGdF_4$: OA	-	Toluene	Solvent evaporation (RT)	AlB_2-type superlattice AB_2-type superlattice $CaTiO_3$-type superlattice	LS_2 LS_2 LS_4	-	240
Cubic $FAPbBr_3$ (5.7)	Spherical $NaGdF_4$ (15.1)	$FAPbBr_3$: - $NaGdF_4$: OA	-	Toluene	Solvent evaporation (RT)	NaCl-type superlattice	LS	-	240
Cubic $CsPbBr_3$ (8.6)	Spherical Fe_3O_4 (19.5)	$CsPbBr_3$: DDAB Fe_3O_4: OA	0.420	Toluene	Solvent evaporation (RT)	NaCl-type superlattices AlB_2-type superlattices $CaTiO_3$-type superlattice	LS LS_2 LS_4	-	242
Spherical β-$NaGdF_4$ (9)	GdF_3 nanoplates: ellipsoidal (16 nm by 10 nm) or rhombic (35 nm by 25 nm)	-	-	Hexane (on DEG)	Air-liquid interface	The nanoplates stack face-to-face and edge-on, forming an ordered lamellar structure with their long axes aligned parallel to the substrate. The β-$NaGdF_4$ nanocrystals fill the gaps between the stacked nanoplates	-	-	238
Spherical Fe_3O_4 (11.0)	$NaYF_4$ nanorods (38.5 nm in length and 19.5 nm in diameter)	-	-	Hexane (on DEG)	Air-liquid interface	2D LS_2 superlattice	LS_2	-	264
Gd_2O_3 nanoplates (23 nm arm length, 7 nm width, and 2 nm thick)	GdF_3 nanoplates (24 nm base, 20 nm height, and 34 nm long axis)	-	-	Hexane (on DEG)	Air-liquid interface	The rhombic nanoplates stand upright between the tripodal ones. Within each 1D string of rhombic and tripodal nanoplates, the nanoplates self-assemble into separate, single-component 1D arrays through shape-specific face-to-face interactions	-	-	237
CdSe/CdS nanorods (15.1 nm in length and 3.8 nm in diameter)	LaF_3 nanodisks (13.2 nm in diameter and 1.6 nm in thickness)	Alkyl-functionalized	-	Hexane (on DEG)	Air-liquid interface	LS-type superlattice (*C2mm* symmetry)	LS	-	235
CdSe/CdS nanorods (15.1 nm in length and 3.8 nm in diameter)	LaF_3 nanodisks (22.3 nm in diameter and 1.6 nm in thickness)	Alkyl-functionalized	-	Hexane (on DEG)	Air-liquid interface	LS_2-type superlattice (*P4mm* symmetry) LS_6-type superlattice (*P4mm* symmetry)	LS_2 LS_6	-	235
Spherical Fe_3O_4 (25)	CdSe/CdS octapods (48 nm in length and 12 nm in diameter) (L/D = 4)	Fe_3O_4: OA Octapods: DDT	0.52[A]	Hexane (on DEG)	Air-liquid interface	Tic-tac-toe arrangement	-	-	270
Cubic $CsPbBr_3$ (8.6)	Spherical Fe_3O_4 (19.8)	$CsPbBr_3$: DDAB Fe_3O_4: OA	0.414	Dodecane (on glyceryl triacetate)	Air-liquid interface	2D LS-type superlattice	LS	-	240

Table 2. Binary superstructures formed through the co-assembly of anisotropic nanoparticles (continuation).

Cubic $CsPbBr_3$ (8.6)	Spherical Fe_3O_4 (19.8)	$CsPbBr_3$: DDAB Fe_3O_4: OA	0.414	Decane (on glyceryl triacetate)	Air-liquid interface	NaCl-type superlattice AlB_2-type superlattice	LS LS_2	-	240
Spherical Pd (5.8)	Octahedral c-In_2O_3 (15.9 nm in diagonal length)	Pd: DDT c-In_2O_3: OA and OAM	-	Toluene	Electrostatic	-	-	-	50
Cubic Au (47)	Cubic Au (from 47 to 85)	DNA	-	water	DNA-guided	NaCl-type superlattice	LS	1:1	289
Cubic Au (52)	Disk-shaped Au (35)	DNA	-	water	DNA-guided	NaCl-type superlattice	LS	1:3	289
Cuboctahedral Au (61 or 95)	Octahedral Au (61 or 95)	Flexible DNA	-	Water	DNA-guided	*rch* superlattice	-	-	306
Tetrahedral Au (82)	Octahedral Au (82)	Flexible DNA	-	Water	DNA-guided	*toh* superlattice	-	-	306
Octahedral Au (78)	Bitetrahedral Au (78)	Flexible DNA	-	Water	DNA-guided	*gtoh* superlattice	-	-	306
Decahedral Au (85)	Octahedral Au (85)	Flexible DNA	-	Water	DNA-guided	P1 quasi-space-filling superlattice	-	-	306
Cubic $CsPbBr_3$ (8.6)	Spherical $NaGdF_4$ (18.6)	$CsPbBr_3$: DDAB $NaGdF_4$: OA	0.439	Toluene	Emulsion-assisted (Toluene-in-FC-40, Toluene-in-HFE-750 surfactant: 008-FS)	$CaTiO_3$-type superlattice	LS_4	-	240

Notation: (ϰ) The size refers to that of the core particles; (†) The size ratio (γ) is the effective size ratio; (ʌ) sphere diameter to octapod length; DDT, dodecanethiol; OA, oleic acid; TOPO, tri-n-octylphosphine oxide; TCE, tetrachloroethylene; RT, room temperature; DDAB, didodecyldimethylammonium bromide; DEG, diethylene glycol; OAM, oleylamine; DNA, deoxyribonucleic acid; c-In_2O_3, cubic-phase In_2O_3; *rch*, rectified cubic honeycomb superlattice; *toh*, tetra-octa honeycomb superlattice; *gtoh*, gyrated tetra-octa honeycomb superlattice.

Table 3. Ternary superstructures formed through the co-assembly of nanoparticles.

Small particles (S) (size in nm)(ϰ)	Large particles (M) (size in nm)(ϰ)	Large particles (L) (size in nm)(ϰ)	Particle functionalization	Size ratio ($\gamma_{S/M}$)(†)	Size ratio ($\gamma_{M/L}$)(†)	Solvent	Co-assembly approach	Superlattice	Stoichiometry	Ref.
Cubic $CsPbBr_3$ (8.6)	Truncated cuboidal PbS (11.7)	Spherical Fe_3O_4 (21.5)	$CsPbBr_3$: DDAB PbS: OA Fe_3O_4: OA	-	-	Toluene	Solvent evaporation (RT)	$CaTiO_3$-type superlattice	LMS_3	240
Cubic $CsPbBr_3$ (8.6)	Truncated cuboidal PbS (11.7)	Spherical Fe_3O_4 (25.1)	$CsPbBr_3$: DDAB PbS: OA Fe_3O_4: OA	-	-	Toluene	Solvent evaporation (RT)	$CaTiO_3$-type superlattice	LMS_3	240
Spherical CdSe (5.8)	Spherical PbSe (7.9)	Spherical PbSe (12.1)	-	-	-	Toluene	Solvent evaporation (70 °C)	AlB_2-type superlattice $MgZn_2$-type superlattice $AlMgB_4$-type superlattice	LS_2 MS_2 LMS_4	160
Cubic $CsPbBr_3$ (8.6)	Truncated cubic PbS (10.7)	Spherical Fe_3O_4 (19.8)	$CsPbBr_3$: DDAB PbS and Fe_3O_4: OA	0.777	0.533	Toluene	Solvent evaporation (RT)	$CaTiO_3$-type superlattice	$LM_xS_3S_{1-x}$	242
Cubic $CsPbBr_3$ (8.6)	Truncated cubic PbS (11.7)	Spherical Fe_3O_4 (25.1)	$CsPbBr_3$: DDAB PbS and Fe_3O_4: OA	0.711	0.46	Toluene	Solvent evaporation (RT)	$CaTiO_3$-type superlattice	LMS_3	242
Spherical FePt (5.0)	Spherical Fe_3O_4 (7.0)	Spherical Fe_3O_4 (16.5)	OA and/or OAM	0.77	0.48	Hexane (on DEG)	Air-liquid interface	2D LMS_2-type superlattice	LMS_2	253
CdSe/CdS nanorods (15.1 nm in length and 3.8 nm in diameter)	LaF_3 nanodisks (13.2 nm in diameter and 1.6 nm in thickness)	LaF_3 nanodisks (22.3 nm in diameter and 1.6 nm in thickness)	Alkyl-functionalized	-	-	Hexane (on DEG)	Air-liquid interface	2D LMS-type superlattice	LMS	235

Notation: (ϰ) The size refers to that of the core particles; (†) The size ratio (γ) is the effective size ratio; DDAB, didodecyldimethylammonium bromide; OA, oleic acid; RT, room temperature; DEG, diethylene glycol; OAM, oleylamine.

Table 4. Binary superstructures formed through the co-assembly of spherical and anisotropic submicrometer-sized particles.

Small particles (S) (size in nm)[×]	Large particles (L) (size in nm)[×]	Particle functionalization	Size ratio (γ)[†]	Solvent	Co-assembly approach	Superlattice	Stoichiometry	$n_L:n_S$	Ref.
Spherical PMMA (372)	Spherical PMMA (642)	Poly-12-hydroxystearic acid	0.58	Decalin and carbon disulphide	Solvent destabilization	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	1:4-6	233
Spherical PMMA (372)	Spherical PMMA (642)	Poly-12-hydroxystearic acid	0.58	Decalin and carbon disulphide	Solvent destabilization	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_{13}	1:9-30	233
Spherical NH_2-PS (60)	Spherical SO_4-PS (368)	-	0.163	Water pH 4-10	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	330
Spherical COOH-PS (59.5)	Spherical SO_4-PS (368)	-	0.162	Water pH 4-10	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	330
Spherical SO_4-PS (70.8)	Spherical SO_4-PS (368)	-	0.192	Water pH 4-10	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	330
Spherical SO_4-PS (70.8)	Spherical NH_2-PS (497.1)	-	0.142	Water pH 4-10	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	330
Spherical NH_2-PS (152.1)	Spherical SO_4-PS (519.8)	-	0.293	Water pH 4-10	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	330
Spherical NH_2-PS (200)	Spherical SO_4-PS (1000)	-	0.2	Water	Solvent evaporation within a confined space (RT, vacuum)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	332
Spherical NH_2-PS (200)	Spherical SO_4-PS (3100)	-	0.065	Water	Solvent evaporation within a confined space (RT, vacuum)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	332
Spherical NH_2-PS (200)	Spherical COOH-PS (2000)	-	0.1	Water	Solvent evaporation within a confined space (RT, vacuum)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	332
Spherical COOH-Si (50)	Spherical NH_2-PS (200)	-	0.25	Water	Solvent evaporation within a confined space (RT, vacuum)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	332
Spherical NH_2-PS (200)	Spherical COOH-Si (2000)	-	0.1	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	333
Spherical NH_2-PS (400)	Spherical COOH-PS (2000)	-	0.2	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_8	-	333

Table 4. Binary superstructures formed through the co-assembly of spherical and anisotropic submicrometer-sized particles (continuation).

Spherical COOH-Si (50)	Spherical NH_2-PS (200)	-	0.25	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	Deviation of LS_8	-	333
Spherical NH_2-PS (110)	Spherical NH_2-PS (200)	-	0.55	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	333
Spherical NH_2-PS (110)	Spherical SO_4-PS (1000)	-	0.11	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	333
Spherical COOH-Si (50)	Spherical SO_4-PS (500)	-	0.05	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	333
Spherical silica (220)	Spherical silica (406)	-	0.54	-	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2 LS_4	-	337
Spherical PS (194)	Spherical silica (406)	-	0.48	-	Solvent evaporation	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2 LS_3	-	337
Spherical NH_2-PS (200)	Spherical SO_4-PS (3100)	-	0.065	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	352
Spherical NH_2-PS (110)	Plain-Si (1000)	-	0.11	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	352
Plain-Si (150)	Plain-Si (1000)	-	0.15	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_8	-	352
Plain-Si (500)	Plain-Si (2000)	-	0.25	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_8	-	352
Plain-Si (722)	Plain-Si (2000)	-	0.36	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_4	-	352
Plain-Si (800)	Plain-Si (2000)	-	0.40	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	$LS_{3.33}$	-	352
Plain-Si (500)	Plain-Si (1000)	-	0.50	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2 LS_3	-	352
Plain-Si (350)	Plain-Si (500)	-	0.70	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS	-	352

Table 4. Binary superstructures formed through the co-assembly of spherical and anisotropic submicrometer-sized particles (continuation).

Plain-PS (722)	Plain-Si (1000)	-	0.72	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS	-	352
Plain-Si (1700)	Plain-Si (2000)	-	0.85	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS	-	352
Spherical NH_2-PS (200)	Spherical SO_4-PS (3100)	-	0.065	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	353
Spherical NH_2-PS (200)	Spherical COOH-PS (2000)	-	0.1	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	353
Spherical COOH-Si (50)	Plain-PS (350)	-	0.14	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	-	353
Spherical NH_2-PS (400)	Spherical COOH-PS (2000)	-	0.2	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_8	-	353
Spherical COOH-Si (50)	Spherical NH_2-PS (200)	-	0.25	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	Deviation of LS_8	-	353
Spherical COOH-Si (50)	Plain-PS (150)	-	0.333	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	353
Spherical SO_4-PS (110)	Spherical NH_2-PS (200)	-	0.55	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	353
Spherical COOH-Si (2000)	Spherical SO_4-PS (3100)	-	0.64	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2 Deviation of LS_2	-	353
Spherical PS (200)	Spherical PS (2000)	-	0.1	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_{20} LS_{26}	-	354
Spherical PS (350)	Spherical PS (2000)	-	0.175	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6 LS_9 LS_{12} LS_{17}	-	354
Spherical PS (500)	Spherical PS (2000)	-	0.25	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6	-	354

Table 4. Binary superstructures formed through the co-assembly of spherical and anisotropic submicrometer-sized particles (continuation).

Spherical PS (130)	Spherical PS (887)	-	0.147	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	2	360
Spherical PS (130)	Spherical PS (887)	-	0.147	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6	4	360
Spherical PS (173)	Spherical PS (887)	-	0.195	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	2	360
Spherical PS (173)	Spherical PS (887)	-	0.195	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6	4	360
Spherical PS (130)	Spherical PS (669)	-	0.194	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	2	360
Spherical PS (130)	Spherical PS (669)		0.194	Water and ethanol (on water)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6	4	360
Spherical PS (202)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.19	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	2	329
Spherical PS (202)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.19	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6	6	329
Spherical PS (202)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.19	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_9	9	329
Spherical PS (225)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.21	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	2	329
Spherical PS (225)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.21	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6	6	329
Spherical PS (225)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.21	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_9	9	329
Spherical PS (303)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.29	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	2	329
Spherical PS (303)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.29	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_6	6	329

Table 4. Binary superstructures formed through the co-assembly of spherical and anisotropic submicrometer-sized particles (continuation).

Spherical PS (336)	Spherical PS (1063)	Carboxylic acid surface functionalization	0.32	Water and ethanol (on water)	Air-liquid interface (Langmuir-Blodgett technique)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	2	329
Spherical PMMA (1980)	Spherical PMMA (2160)		0.92	CHB and cis-decalin	Electrostatic (TBAB to adjust particle charges)	CsCl-type superlattice	LS	1:1	196
Spherical silica (1040)	Spherical PMMA (1160)	-	0.9	CHB and cis-decalin	Electrostatic (TBAB to adjust particle charges)	CsCl-type superlattice	LS	-	196
Spherical PMMA (720)	Spherical PMMA (2320)	-	0.31	CHB and cis-decalin	Electrostatic (TBAB to adjust particle charges)	*bcc*-LS_6-type superlattice	LS_6	1:8	196
Spherical PMMA (720)	Spherical PMMA (2320)	-	0.31	CHB and cis-decalin	Electrostatic (TBAB to adjust particle charges)	NaCl-type superlattice NiCl-type superlattice	LS LS	1:8	196
Spherical PMMA (720)	Spherical PMMA (777)	-	0.93 ± 0.01	CHPB and CD	Electrostatic	CsCl-type superlattice NaCl-type superlattice	LS LS	-	367
Spherical TPM (1000)	Spherical TPM (1000)	Flexible DNA	1	Water	DNA-guided	CsCl-type superlattice	LS	-	257
Spherical TPM (540)	Spherical TPM (1000)	Flexible DNA	0.54	Water	DNA-guided	AlB_2-type superlattice	LS_2	-	257
Spherical TPM (540)	Spherical TPM (1500)	Flexible DNA	0.36	Water	DNA-guided	*bcc*-LS_6-type superlattice	LS_6	-	257
Spherical silica (222)	Spherical silica (442)	-	0.5	Ethanol and ethylene glycol	Spin-coating (3,000 rpm)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	374
Spherical silica (222)	Spherical silica (442)	-	0.5	Ethanol and ethylene glycol	Spin-coating (10,000 rpm)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_3	-	374
Spherical silica (442)	Spherical silica (891)	-	0.5	Ethanol and ethylene glycol	Spin-coating (3,000 rpm)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	374
Spherical silica (519)	Spherical silica (891)	-	0.58	Ethanol and ethylene glycol	Spin-coating (3,000 rpm)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2 LS_3	-	374
Spherical silica (222)	Spherical silica (891)	-	0.25	Ethanol and ethylene glycol	Spin-coating (3,000 rpm)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	374

Table 4. Binary superstructures formed through the co-assembly of spherical and anisotropic submicrometer-sized particles (continuation).

Spherical PS (400)	Spherical PS (1800)	-	0.22	Water	Electric field-assisted (Glass substrate coated with ITO)	Large particles self-assembled into hexagonally close-packed monolayers. Each interstitial site accommodated four small particles arranged in an inverse pyramid, with each large particle surrounded by 24 small ones	LS_8	-	376
Spherical PS (150)	Spherical PS (700)	-	0.2	Water	Electric field-assisted (Glass substrate coated with ITO)	Large particles self-assembled into hexagonally close-packed monolayers. Each interstitial site accommodated four small particles arranged in an inverse pyramid, with each large particle surrounded by 24 small ones.	LS_8	-	376
Spherical PS (400)	Spherical PS (700)	-	0.57	Water	Electric field-assisted (Glass substrate coated with ITO)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	376
Spherical PS (640)	Spherical PS (700)	-	0.91	Water	Electric field-assisted (Glass substrate coated with ITO)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS	-	376
Spherical PS (240)	Spherical PS (700)	-	0. 34	Water	Electric field-assisted (Glass substrate coated with ITO)	LS_2 or LS_4 depending on the number of small particles deposited onto the monolayer of large particles	LS_2 LS_4	-	376
Spherical silica (410)	Spherical silica (1370)	-	0.3	Glycerol and water	Electric field-assisted	NaCl-type superlattices	LS	-	377
Spherical PS (100)	Filamentous fd virus (6.6 nm diameter, 880 nm contour length, 2.2 μm persistence length)	-	-	Water	Solvent destabilization	"Columnar" phase (at low concentration of spheres) " Lamellar" phase (at high concentration of spheres)	-	-	347
Cubic ZIF-8 (135 nm in length)	Spherical PS (600)	Pluronic F127	-	Water	Solvent evaporation (40 °)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	349
Cubic ZIF-8 (196 nm in length)	Spherical PS (600)	Pluronic F127	-	Water	Solvent evaporation (40 °)	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	-	349
Cubic ZIF-8 (196 nm in length)	TRD ZIF-8 (414 nm in length, with a truncation of 0.68)	Cubic ZIF-8: Pluronic F127 TRD ZIF-8: Pluronic F127-COOH	-	Water	Solvent evaporation (40 °)	NaCl-type superlattices	LS	-	349

Table 4. Binary superstructures formed through the co-assembly of spherical and anisotropic submicrometer-sized particles (continuation).

Spherical $NaYF_4$ (32)	Spherical MOF-801 (100)	Oleyl phosphate	0.32	Cyclohexane (on DEG)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	350
Spherical Fe_3O_4 (21)	Spherical MOF-801 (40)	Oleyl phosphate	0.53	Cyclohexane (on DEG)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS	350
Truncated octahedral MOF-801 (40 in length)	Spherical MOF-801 (120)	Oleyl phosphate	0.33	Cyclohexane (on DEG)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	350
Octahedral UiO-66-NH_2 (35 in length)	Spherical MOF-801 (120)	Oleyl phosphate	0.30	Cyclohexane (on DEG)	Air-liquid interface	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LS_2	350

Notation: (ϰ) The size refers to that of the core particles; (†) The size ratio (γ) is the effective size ratio; PMMA, polymethylmethacrylate; PS, polystyrene; RT, room temperature; CHB, cyclohexyl bromide; CHPB, cycloheptyl bromide; CD, cis-decalin; DNA, deoxyribonucleic acid; TPM, 3-(trimethoxysilyl) propyl methacrylate; ITO, indium tin oxide; TRD, truncated rhombic dodecahedral.

Table 5. Ternary and quaternary superstructures formed through the co-assembly of submicrometer-sized particles.

Small particles (S) (size in nm)[ϰ]	Large particles (M) (size in nm)[ϰ]	Large particles (L) (size in nm)[ϰ]	Large particles (XL) (size in nm)[ϰ]	Particle functionalization	Size ratio ($\gamma_{S/M}$)[†]	Size ratio ($\gamma_{M/L}$)[†]	Size ratio ($\gamma_{M/XL}$)[†]	Solvent	Co-assembly approach	Superlattice	Stoichiometry	Ref.
Spherical silica (6)	Spherical PMMA (84)	Spherical PS (465)	-	-	0.071	0.18	-	Water	Solvent destabilization	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	351
Spherical COOH-Si (50)	Spherical NH_2-PS (200)	Spherical COOH-PS (2000)	-	-	0.25	0.1	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	353
Spherical COOH-Si (50)	Spherical NH_2-PS (200)	Spherical SO_4-PS (3100)	-	-	0.25	0.065	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	353
Spherical COOH-Si (50)	Spherical NH_2-PS (200)	Spherical SO_4-PS (1000)	-	-	0.25	0.2	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	353
Spherical COOH-Si (25)	Plain-PS (722)	Plain-Si (5000)	-	-	0.035	0.144	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	353
Spherical COOH-Si (50)	Spherical NH_2-PS (200)	Plain-Si (1000)	Plain-Si (5000)	-	0.25	0.2	0.2	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	353
Spherical COOH-Si (25)	Spherical NH_2-PS (200)	Plain-Si (1000)	Plain-Si (5000)	-	0.125	0.2	0.2	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	353
Plain-Si (150)	Plain-Si (1000)	Plain-Si (2000)	-	-	0.15	0.5	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	352
Plain-Si (150)	Plain-Si (500)	Plain-Si (1000)	-	-	0.30	0.5	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	-	352
Plain-PS (800)	Plain-Si (500)	Plain-Si (1000)	-	-	0.63	0.5	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LMS_3	352
Plain-Si (1700)	Plain-Si (1000)	Plain-Si (2000)	-	-	0.59	0.5	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LMS_4	352
Plain-Si (500)	Plain-Si (1000)	Plain-Si (2000)	-	-	0.5	0.5	-	Water	Solvent evaporation within a confined space	2D superlattice: large particles form a hexagonal pattern, with small particles occupying the spaces	LM_2S_7 LM_2S_{10} LM_2S_{11}	352
Spherical PS (150)	Spherical PS (240)	Spherical PS (700)	-	-	0.625	0.34	-	Water	Electric field-assisted (Glass substrate coated with ITO)	Five 150 nm PS particles filled each secondary interstitial site between two 240 nm PS particles, with one particle positioned in the middle and four others forming a layer above it	LM_2S_{15}	376

Notation: (ϰ) The size refers to that of the core particles; (†) The size ratio (γ) is the effective size ratio; PMMA, polymethylmethacrylate; PS, polystyrene; ITO, indium tin oxide.

References**

** The number of references in the Supporting Information matches those in the main manuscript.

[24] C. J. Kiely, J. Fink, M. Brust, D. Bethell, D. J. Schiffrin, Nature 396 (1998) 444-446. https://doi.org/10.1038/24726.
[29] E. V. Shevchenko, D. V. Talapin, S. O'Brien, C. B. Murray, J. Am. Chem. Soc. 127 (2005) 8741-8747. https://doi.org/10.1021/ja050980i.
[31] E. V. Shevchenko, D. V. Talapin, N. A. Kotov, S. O'Brien, C. B. Murray, Nature 439 (2006) 55-59. https://doi.org/10.1038/nature04414.
[32] E. V. Shevchenko, D. V. Talapin, C. B. Murray, S. O'Brien, J. Am. Chem. Soc. 128 (2006) 3620-3637. https://doi.org/10.1021/ja0564261.
[33] D. V. Talapin, E. V. Shevchenko, M. I. Bodnarchuk, X. Ye, J. Chen, C. B. Murray, Nature 461 (2009) 964-967. https://doi.org/10.1038/nature08439.
[37] Z. Chen, J. Moore, G. Radtke, H. Sirringhaus, S. O'Brien, J. Am. Chem. Soc. 129 (2007) 15702-15709. https://doi.org/10.1021/ja0756239.
[38] Z. Chen, S. O'Brien, ACS Nano 2 (2008) 1219-1229. https://doi.org/10.1021/nn800187v.
[41] M. I. Bodnarchuk, M. V. Kovalenko, W. Heiss, D. V. Talapin, J. Am. Chem. Soc. 132 (2010) 11967-11977. https://doi.org/10.1021/ja103083q.
[49] A. M. Kalsin, M. Fialkowski, M. Paszewski, S. K. Smoukov, K. J. M. Bishop, B. A. Grzybowski, Science 312 (2006) 420-424. https://doi.org/10.1126/science.1125124.
[50] Z. Sun, Z. Luo, J. Fang, ACS Nano 4 (2010) 1821-1828. https://doi.org/10.1021/nn100302v.
[149] M. I. Bodnarchuk, L. Li, A. Fok, S. Nachtergaele, R. F. Ismagilov, D. V. Talapin, J. Am. Chem. Soc. 133 (2011) 8956-8960. https://doi.org/10.1021/ja201129n.
[160] W. H. Evers, H. Friedrich, L. Filion, M. Dijkstra, D. Vanmaekelbergh, Angew. Chem. Int. Ed. 48 (2009) 9655-9657. https://doi.org/10.1002/anie.200904821.
[181] D. K. Smith, B. Goodfellow, D. M. Smilgies, B. A. Korgel, J. Am. Chem. Soc. 131 (2009) 3281-3290. https://doi.org/10.1021/ja8085438.
[184] F. X. Redl, K. S. Cho, C. B. Murray, S. O'Brien, Nature 423 (2003) 968-971. https://doi.org/10.1038/nature01702.
[190] A. E. Saunders, B. A. Korgel, ChemPhysChem 6 (2005) 61-65. https://doi.org/10.1002/cphc.200400382.

[194] X. Ye, J. Chen, C. B. Murray, J. Am. Chem. Soc. 133 (2011) 2613-2620. https://doi.org/10.1021/ja108708v.
[196] M. E. Leunissen, C. G. Christova, A.-P. Hynninen, C. P. Royall, A. I. Campbell, A. Imhof, M. Dijkstra, R. van Roij, A. van Blaaderen, Nature 437 (2005) 235-240. https://doi.org/10.1038/nature03946.
[206] M. I. Bodnarchuk, R. Erni, F. Krumeich, M. V. Kovalenko, Nano Lett. 13 (2013) 1699-1705. https://doi.org/10.1021/nl4002475.
[208] Z. Yang, J. Wei, P. Bonville, M.-P. Pileni, J. Am. Chem. Soc. 137 (2015) 4487-4493. https://doi.org/10.1021/jacs.5b00332.
[211] K. Overgaag, W. Evers, B. de Nijs, R. Koole, J. Meeldijk, D. Vanmaekelbergh, J. Am. Chem. Soc. 130 (2008) 7833-7835. https://doi.org/10.1021/ja802932m.
[214] M. A. Boles, D. V. Talapin, J. Am. Chem. Soc. 137 (2015) 4494-4502. https://doi.org/10.1021/jacs.5b00839.
[217] J. Wei, N. Schaeffer, M.-P. Pileni, J. Am. Chem. Soc. 137 (2015) 14773-14784. https://doi.org/10.1021/jacs.5b09959.
[221] I. Coropceanu, M. A. Boles, D. V. Talapin, J. Am. Chem. Soc. 141 (2019) 5728-5740. https://doi.org/10.1021/jacs.8b12539.
[233] P. Bartlett, R. H. Ottewill, P. N. Pusey, Phys. Rev. Lett. 68 (1992) 3801-3804. https://doi.org/10.1103/PhysRevLett.68.3801.
[234] I. Cherniukh, G. Rainò, T. V. Sekh, C. Zhu, Y. Shynkarenko, R. A. John, E. Kobiyama, R. F. Mahrt, T. Stöferle, R. Erni, M. V. Kovalenko, M. I. Bodnarchuk, ACS Nano 15 (2021) 16488-16500. https://doi.org/10.1021/acsnano.1c06047.
[235] T. Paik, B. T. Diroll, C. R. Kagan, C. B. Murray, J. Am. Chem. Soc. 137 (2015) 6662-6669. https://doi.org/10.1021/jacs.5b03234.
[237] T. Paik, C. B. Murray, Nano Lett. 13 (2013) 2952-2956. https://doi.org/10.1021/nl401370n.
[238] T. Paik, D.-K. Ko, T. R. Gordon, V. Doan-Nguyen, C. B. Murray, ACS Nano 5 (2011) 8322-8330. https://doi.org/10.1021/nn203049t.
[240] I. Cherniukh, T. V. Sekh, G. Rainò, O. J. Ashton, M. Burian, A. Travesset, M. Athanasiou, A. Manoli, R. A. John, M. Svyrydenko, V. Morad, Y. Shynkarenko, F. Montanarella, D. Naumenko, H. Amenitsch, G. Itskos, R. F. Mahrt, T. Stöferle, R. Erni, M. V. Kovalenko, M. I. Bodnarchuk, ACS Nano 16 (2022) 7210-7232 https://doi.org/10.1021/acsnano.1c10702.
[241] E. V. Shevchenko, J. B. Kortright, D. V. Talapin, S. Aloni, A. P. Alivisatos, Adv. Mater. 19 (2007) 4183-4187. https://doi.org/10.1002/adma.200701470.

[242] I. Cherniukh, G. Rainò, T. Stöferle, M. Burian, A. Travesset, D. Naumenko, H. Amenitsch, R. Erni, R. F. Mahrt, M. I. Bodnarchuk, M. V. Kovalenko, Nature 593 (2021) 535-542. https://doi.org/10.1038/s41586-021-03492-5.

[248] A. Dong, J. Chen, P. M. Vora, J. M. Kikkawa, C. B. Murray, Nature 466 (2010) 474-477. https://doi.org/10.1038/nature09188.

[250] T. Kister, M. Mravlak, T. Schilling, T. Kraus, Nanoscale 8 (2016) 13377-13384. https://doi.org/10.1039/C6NR01940D.

[253] A. Dong, X. Ye, J. Chen, C. B. Murray, Nano Lett. 11 (2011) 1804-1809. https://doi.org/10.1021/nl200468p.

[254] X. Ye, C. Zhu, P. Ercius, S. N. Raja, B. He, M. R. Jones, M. R. Hauwiller, Y. Liu, T. Xu, A. P. Alivisatos, Nat. Commun. 6 (2015) 10052. https://doi.org/10.1038/ncomms10052.

[256] R. J. Macfarlane, B. Lee, M. R. Jones, N. Harris, G. C. Schatz, C. A. Mirkin, Science 334 (2011) 204-208. https://doi.org/10.1126/science.1210493.

[257] Y. Wang, Y. Wang, X. Zheng, É. Ducrot, J. S. Yodh, M. Weck, D. J. Pine, Nat. Commun. 6 (2015) 7253. https://doi.org/10.1038/ncomms8253.

[264] X. Ye, J. A. Millan, M. Engel, J. Chen, B. T. Diroll, S. C. Glotzer, C. B. Murray, Nano Lett. 13 (2013) 4980-4988. https://doi.org/10.1021/nl403149u.

[270] A. Castelli, J. de Graaf, M. Prato, L. Manna, M. P. Arciniegas, ACS Nano 10 (2016) 4345-4353. https://doi.org/10.1021/acsnano.5b08018.

[278] D. Nykypanchuk, M. M. Maye, D. Van Der Lelie, O. Gang, Nature 451 (2008) 549-552. https://doi.org/10.1038/nature06560.

[289] M. N. O'Brien, M. R. Jones, B. Lee, C. A. Mirkin, Nat. Mater. 14 (2015) 833-839. https://doi.org/10.1038/nmat4293.

[306] W. Zhou, Y. Li, K. Je, T. Vo, H. Lin, B. E. Partridge, Z. Huang, S. C. Glotzer, C. A. Mirkin, Science 383 (2024) 312-319. https://doi.org/10.1126/science.adj1021.

[308] Y. Wang, J. Chen, C. Zhu, B. Zhu, S. Jeong, Y. Yi, Y. Liu, J. Fiadorwu, P. He, X. Ye, Nano Lett. 21 (2021) 5053-5059. https://doi.org/10.1021/acs.nanolett.1c00890.

[315] P.-p. Wang, Q. Qiao, Y. Zhu, M. Ouyang, J. Am. Chem. Soc. 140 (2018) 9095-9098. https://doi.org/10.1021/jacs.8b05643.

[316] A. Travesset, Proc. Natl. Acad. Sci. 112 (2015) 9563-9567. https://doi.org/10.1073/pnas.1504677112.

[329] N. Vogel, L. de Viguerie, U. Jonas, C. K. Weiss, K. Landfester, Adv. Funct. Mater. 21 (2011) 3064-3073. https://doi.org/10.1002/adfm.201100414.

[330] R. Mukhopadhyay, O. Al-Hanbali, S. Pillai, A. G. Hemmersam, R. L. Meyer, A. C. Hunter, K. J. Rutt, F. Besenbacher, S. M. Moghimi, P. Kingshott, J. Am. Chem. Soc. 129 (2007) 13390-13391. https://doi.org/10.1021/ja075988c.

[332] G. Singh, S. Pillai, A. Arpanaei, P. Kingshott, Adv. Mater. 23 (2011) 1519-1523. https://doi.org/10.1002/adma.201004657.

[333] G. Singh, H. J. Griesser, K. Bremmell, P. Kingshott, Adv. Funct. Mater. 21 (2011) 540-546. https://doi.org/10.1002/adfm.201001340.

[337] K. P. Velikov, C. G. Christova, R. P. A. Dullens, A. van Blaaderen, Science 296 (2002) 106-109. https://doi.org/10.1126/science.1067141.

[347] M. Adams, Z. Dogic, S. L. Keller, S. Fraden, Nature 393 (1998) 349-352. https://doi.org/10.1038/30700.

[349] L. Meng, J. Fonseca, R. Sánchez-Naya, A. M. Ghadiri, I. Imaz, D. Maspoch, J. Am. Chem. Soc. 146 (2024) 21225-21230. https://doi.org/10.1021/jacs.4c07194.

[350] X. Wu, Y. Gao, H. Wang, Z. Zhang, X. Xi, D. Yang, T. Li, A. Dong, J. Am. Chem. Soc. 147 (2025) 6361-6366. https://doi.org/10.1021/jacs.4c17724.

[351] J. Wang, Q. Li, W. Knoll, U. Jonas, J. Am. Chem. Soc. 128 (2006) 15606-15607. https://doi.org/10.1021/ja067221a.

[352] G. Singh, S. Pillai, A. Arpanaei, P. Kingshott, Adv. Funct. Mater. 21 (2011) 2556-2563. https://doi.org/10.1002/adfm.201002716.

[353] G. Singh, S. Pillai, A. Arpanaei, P. Kingshott, Soft Matter 7 (2011) 3290-3294. https://doi.org/10.1039/C0SM01360A.

[354] Z. Dai, Y. Li, G. Duan, L. Jia, W. Cai, ACS Nano 6 (2012) 6706-6716. https://doi.org/10.1021/nn3013178.

[360] J. Yu, Q. Yan, D. Shen, ACS Appl. Mater. Interfaces 2 (2010) 1922-1926. https://doi.org/10.1021/am100250c.

[367] P. Bartlett, A. I. Campbell, Phys. Rev. Lett. 95 (2005) 128302. https://doi.org/10.1103/PhysRevLett.95.128302.

[374] D. Wang, H. Möhwald, Adv. Mater. 16 (2004) 244-247. https://doi.org/10.1002/adma.200305565.

[376] X. Huang, J. Zhou, M. Fu, B. Li, Y. Wang, Q. Zhao, Z. Yang, Q. Xie, L. Li, Langmuir 23 (2007) 8695-8698. https://doi.org/10.1021/la700512j.

[377] E. C. M. Vermolen, A. Kuijk, L. C. Filion, M. Hermes, J. H. J. Thijssen, M. Dijkstra, A. van Blaaderen, Proc. Natl. Acad. Sci. 106 (2009) 16063-16067. https://doi.org/10.1073/pnas.0900605106.

[406] E. V. Shevchenko, D. V. Talapin, A. L. Rogach, A. Kornowski, M. Haase, H. Weller, J. Am. Chem. Soc. 124 (2002) 11480-11485. https://doi.org/10.1021/ja025976l.