

# **Supporting information for: Phase Diagram and Structure Map of Binary Nanoparticle Superlattices from a Lennard-Jones Model**

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# List and maps of structures searched by genetic algorithm

Table S1: Table of Stable Structures

Structure	$\gamma = \frac{\sigma_{BB}}{\sigma_{AA}}$	Space Group	Motif	$\frac{a,b,c}{\alpha, \beta, \gamma}$	Wyckoff Position
<i>MgZn<sub>2</sub></i> <sup>(194)</sup>	0.8 0.9	194	Motif-16-2	2.0 4.8 2.0	A(4f) 0.333 0.667 0.437
				129.0 60.0 129.1	B(2a) 0.0 0.0 0.0
					B(6h) 0.831 0.663 0.25
<i>AlB<sub>2</sub></i> <sup>(191)</sup>	0.5 0.6 0.7	191	Motif-12-3	7.0 1.9 3.8	A(1a) 0.0 0.0 0.0
				137.8 42.6 158.8	B(2d) 0.333 0.667 0.5
<i>MoB</i> <sup>(141)</sup>	0.7	141	Motif-13-1	1.8 1.8 4.8	A(8e) 0.0 0.25 0.574
				79.4 79.3 90.0	B(8e) 0.0 0.25 0.033
<i>CaB<sub>6</sub></i> <sup>(221)</sup>	0.4	221	Motif-24-2	3.7 2.6 2.6	A(1a) 0.0 0.0 0.0
				90.0 90.0 135.0	B(6f) 0.21 0.5 0.5
<i>NaCl</i> <sup>(225)</sup>	0.4 0.5	225	Motif-6-4	4.7 2.7 2.7	A(4a) 0.0 0.0 0.0
				90.0 73.2 150.0	B(4b) 0.5 0.5 0.5
<i>ThSi<sub>2</sub></i> <sup>(141)</sup>	0.5	141	Motif-12-2	3.1 2.2 4.0	A(4b) 0.0 0.25 0.375
				105.5 67.8 135.0	B(8e) 0.0 0.25 -0.041
<i>NaZn<sub>13</sub></i> <sup>(226)</sup>	0.6	226	Motif-24-4	12.3 12.3 12.3	A(8a) 0.250 0.250 0.250
				90 90 90	B(96i) 0.000 0.620 0.320
					B(8b) 0.000 0.000 0.000
<i>A<sub>4</sub>B<sub>16</sub></i> <sup>(127)</sup>	0.5	127	Motif-18-1		A(4g) 0.319 0.819 0.0
				4.3 6.5 2.3	B(8j) 0.324 0.549 0.5
				110.6 90.0 131.4	B(4h) 0.089 0.589 0.5
					B(4e) 0.0 0.0 0.783

					A(4i) 0.115 0.0 0.851
					A(2c) 0.0 0.0 0.5
$PuNi_4^{(12)}$	0.7 0.8	12	Motif-18-3 Motif-16-2	2.1 4.5 2.1 66.9 60.0 85.2	B(8j) 0.132 0.331 -0.095 B(8j) 0.814 0.25 0.696 B(4h) 0.0 0.667 0.5 B(4i) 0.435 0.0 0.304
$dhcp^{(194)}$	0.3	194	Motif-6-3	7.3 5.9 3.1 105.1 50.9 125.8	A(2c) 0.333 0.667 0.25 B(2a) 0.0 0.0 0.0
$AB_4_{(2)}^{(139)}$ or $A_4B_{16}_{(2)}^{(139)}$	0.6 0.7	139	Motif-18-2	2.6 1.9 7.5 90.0 68.6 111.4	A(2a) 0.0 0.0 0.0 B(4d) 0.0 0.5 0.25 B(4e) 0.0 0.0 0.354
$CaCu_5^{(191)}$	0.7	191	Motif-18-3	4.2 2.8 1.6 125.1 67.9 150.8	A(1a) 0.0 0.0 0.0 B(3g) 0.5 0.0 0.5 B(2c) 0.333 0.667 0.0
$TePt^{(166)}$	0.5	166	Motif-6-2	4.7 2.2 4.1 75.0 27.4 90.0	A(6c) 0.0 0.0 0.092 B(6c) 0.0 0.0 0.667
$PuNi_3^{(166)}$	0.8	166	Motif-16-2 Motif-18-3	2.1 4.1 2.1 90.0 60.0 121.3	A(6c) 0.0 0.0 0.859 A(3a) 0.0 0.0 0.0 B(18h) 0.166 0.834 0.253 B(6c) 0.0 0.0 0.334 B(3b) 0.0 0.0 0.5
$CrB^{(63)}$	0.6 0.7	63	Motif-13-1	1.9 2.7 2.9 103.8 70.5 135.6	A(4c) 0.0 0.358 0.25 B(4c) 0.0 0.056 0.25
$TiCu_3^{(59)}$	0.6 0.7	59	Motif-12-4	4.3 2.5 1.7 90.0 35.0 90.0	A(2a) 0.25 0.25 0.11 B(4e) 0.25 0.035 0.612 B(2b) 0.25 0.75 0.297

$A_2B_{12}^{(139)}$	0.3	139	Motif-24-1	3.9 3.1 4.9 101.1 90.0 128.3	A(2a) 0.0 0.0 0.0 B(8j) 0.138 0.5 0.0 B(4d) 0.0 0.5 0.25
$KHg_2^{(74)}$	0.7	74	Motif-12-5	2.4 2.4 2.4 101.7 94.6 135.8	A(4e) 0.0 0.25 0.446 B(8h) 0.0 -0.061 0.169
$A_2B_{18}^{(194)}$	0.4	194	Motif-30-2	3.6 5.2 3.6 110.6 60.0 134.8	A(2c) 0.333 0.667 0.25 B(12k) -0.098 0.804 0.111 B(6h) 0.76 0.52 0.25
$A_4B_6^{(15)}$	0.7	15	Motif-15-3	2.5 3.2 2.5 78.1 119.6 102.0	A(8f) 0.153 0.384 0.15 B(4e) 0.0 0.822 0.25 B(8f) 0.883 0.114 0.009
$A_2B_6^{(63)}$	0.6 0.7	63	Motif-15-4	1.7 3.9 9.4 150.0 22.6 153.2	A(4c) 0.0 0.895 0.25 B(4c) 0.0 0.679 0.25 B(4c) 0.0 0.25 0.25 B(4c) 0.0 0.536 0.25
$Mg_3Cd^{(194)}$	0.5	194	Motif-12-4	3.3 3.3 6.9 118.4 17.5 120.0	A(2c) 0.333 0.667 0.25 B(6h) 0.891 0.782 0.25
$A_2B_4^{(227)}$	0.9	227	Motif-16-2	2.1 2.9 2.1 135.0 60.0 135.0	A(8a) 0.125 0.125 0.125 B(16d) 0.5 0.5 0.5

					A(4i) 0.834 0.0 -0.096
					A(4i) 0.332 0.0 0.692
					A(2c) 0.0 0.0 0.5
					B(4i) 0.153 0.0 0.826
$A_5B_{15}^{(12)}_{(10)}$	0.3	12	Motif-6-4	3.1 3.1 13.4	B(4i) -0.013 0.0 0.23
			Motif-15-2	97.2 76.2 121.2	B(8j) 0.083 0.252 0.798
					B(4i) 0.655 0.0 0.769
					B(4i) 0.489 0.0 0.172
					B(4i) 0.667 0.0 0.596
					B(2b) 0.0 0.5 0.0
					A(4i) 0.128 0.0 0.89
					B(4i) 0.531 0.0 0.595
					B(8j) 0.222 0.295 0.396
$A_2B_{18}^{(12)}_{(1)}$	0.3	12	Motif-30-1	5.7 4.2 2.9	B(8j) -0.025 0.277 0.624
				134.6 104.7 81.3	B(4i) 0.486 0.0 0.85
					B(4i) 0.338 0.0 0.598
					B(4i) 0.262 0.0 0.381
					B(4i) 0.582 0.0 0.359
$Zr_2Cu^{(139)}$	0.8	139	Motif-14-1	3.8 4.7 4.4	A(4e) 0.0 0.0 0.851
				147.6 45.6 165.7	B(2b) 0.0 0.0 0.5
					A(4f) 0.333 0.667 0.887
$BaCu^{(194)}$	0.5	194	Motif-6-2	3.7 2.2 13.9	B(2c) 0.333 0.667 0.25
				81.0 36.1 90.0	B(2b) 0.0 0.0 0.25
					A(4a) 0.0 0.0 0.0
$AB_9^{(216)}_{(1)}$	0.5	216	Motif-28-1	3.9 2.8 2.8	B(16e) 0.402 0.402 0.402
				120.2 44.9 135.1	B(16e) 0.655 0.655 0.655
					B(4c) 0.25 0.25 0.25

$A_8B_4^{(12)}_{(12)}$	0.5	12	Motif-3-1	4.2 8.8 4.2 133.7 62.9 145.1	A(4i) -0.082 0.0 0.219 A(4i) 0.344 0.0 0.693 B(4i) 0.738 0.0 -0.089
$A_4B_6^{(166)}_{(9)}$	0.5	166	Motif-6-2 Motif-12-3	2.1 2.1 8.7 90.1 75.7 59.9	A(6c) 0.0 0.0 0.631 A(6c) 0.0 0.0 0.456 B(6c) 0.0 0.0 0.079 B(6c) 0.0 0.0 0.746 B(6c) 0.0 0.0 0.167
$A_2B_{18}^{(71)}_{(1)}$	0.3	71	Motif-21-3	5.6 3.2 2.9 90.0 75.0 106.5	A(4i) 0.0 0.0 0.851 B(4j) 0.5 0.0 0.544 B(8m) 0.17 0.0 0.426 B(8l) 0.0 0.242 0.454 B(4i) 0.0 0.0 0.348 B(8n) 0.769 0.652 0.0 B(2c) 0.5 0.5 0.0 B(2b) 0.0 0.5 0.5
$A_2B_{12}^{(11)}_{(1)}$	0.6	11	Motif-20-1	4.6 3.6 2.0 107.1 76.4 143.2	A(2e) 0.896 0.25 0.778 B(4f) 0.318 0.562 0.606 B(2e) 0.831 0.25 0.174 B(2b) 0.5 0.0 0.0 B(2e) 0.21 0.25 0.438 B(2e) 0.331 0.25 0.172

					A(2i) 0.18 0.218 0.243
					B(2i) 0.326 0.826 -0.003
					B(2i) -0.04 0.332 0.589
					B(2i) 0.339 0.346 0.567
$A_2B_{18}^{(2)}$	0.5	2	Motif-28-2	3.0 4.0 3.0 92.9 70.8 111.1	B(2i) 0.645 0.795 0.147 B(2i) -0.006 0.839 0.122 B(2i) 0.609 0.456 0.124 B(2i) 0.337 0.696 0.581 B(2i) 0.256 -0.005 0.702 B(2i) 0.629 0.364 0.708
$Mg_2Si_3^{(12)}$	0.7	12	Motif-15-5	2.4 6.5 2.4 117.6 44.3 141.7	A(4i) -0.005 0.0 0.173 A(4i) 0.196 0.0 0.607 B(4i) 0.644 0.0 0.059 B(4i) 0.275 0.0 0.147 B(4i) 0.509 0.0 0.386
$A_4B_4^{(62)}$	0.5	62	Motif-6-5	4.1 4.2 4.7 76.0 27.7 90.0	A(4c) 0.181 0.25 0.342 B(4c) 0.523 0.25 0.583
$A_2B_{10}^{(63)}$	0.6	63	Motif-18-2	2.7 1.9 6.8 114.9 46.2 135.1	A(4c) 0.0 0.362 0.25 B(4c) 0.0 0.2 0.25 B(4c) 0.0 -0.026 0.25 B(4c) 0.0 0.75 0.25 B(4c) 0.0 0.082 0.25 B(4c) 0.0 0.523 0.25
$A_2B_8^{(139)}$	0.6	139	Motif-16-1	2.9 2.9 2.7 117.7 62.3 124.6	A(4e) 0.0 0.0 0.777 B(8h) 0.289 0.289 0.0 B(8g) 0.0 0.5 0.623

					A(2a) 0.0 0.0 0.0
					A(2a) 0.794 0.0 0.41
					B(2a) 0.739 0.0 0.706
					B(2a) 0.195 0.0 0.596
					B(4b) 0.297 0.192 0.705
					B(2a) 0.436 0.0 0.78
					B(2a) 0.105 0.0 0.705
$A_2B_{18}^{(160)}_{(1)}$	0.3	160	Motif-21-4	3.0 3.0 6.3 90.4 76.1 119.9	B(4b) -0.02 0.241 0.705 B(4b) 0.118 0.265 0.63
					B(2a) 0.514 0.0 0.557
					B(2a) 0.383 0.0 0.63
					B(4b) 0.171 0.265 0.78
					B(2a) 0.617 0.0 0.853
					B(2a) 0.566 0.0 0.705
					B(2a) 0.271 0.0 0.814
					B(2a) 0.398 0.0 0.205
$A_2B_6^{(139)}_{(13)}$	0.4	139	Motif-12-1	3.8 4.9 2.7 105.9 45.0 112.8	A(4e) 0.0 0.0 0.144 B(8j) 0.209 0.5 0.0 B(4e) 0.0 0.0 0.411
$A_4B_2^{(12)}_{(11)}$	0.5 0.6	12	Motif-3-1 Motif-15-1	3.5 3.0 6.4 146.3 38.5 148.0	A(4i) 0.654 0.0 0.82 A(4i) 0.077 0.0 0.283 B(4i) 0.736 0.0 0.402
$A_2B_{18}^{(15)}_{(1)}$	0.6	15	Motif-25-1	2.7 4.2 2.7 93.5 80.3 127.5	A(4e) 0.0 0.203 0.25 B(8f) 0.795 0.095 0.889 B(8f) 0.164 0.388 0.639 B(8f) 0.205 0.137 -0.084
					B(8f) 0.085 0.428 -0.087
					B(4e) 0.0 0.799 0.25

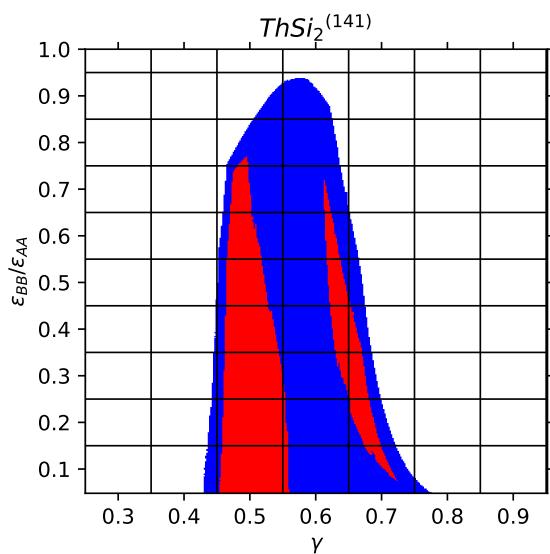
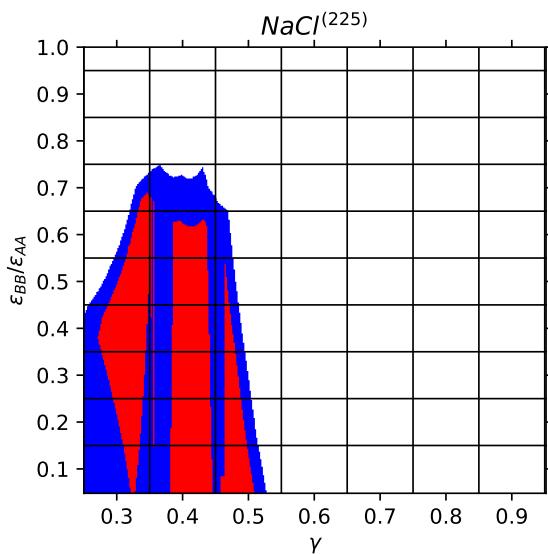
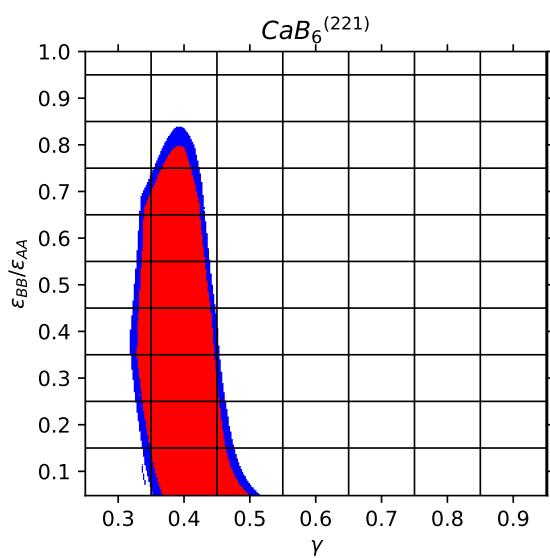
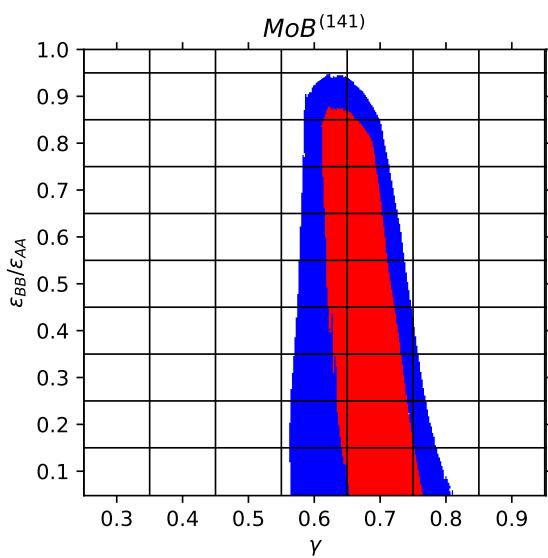
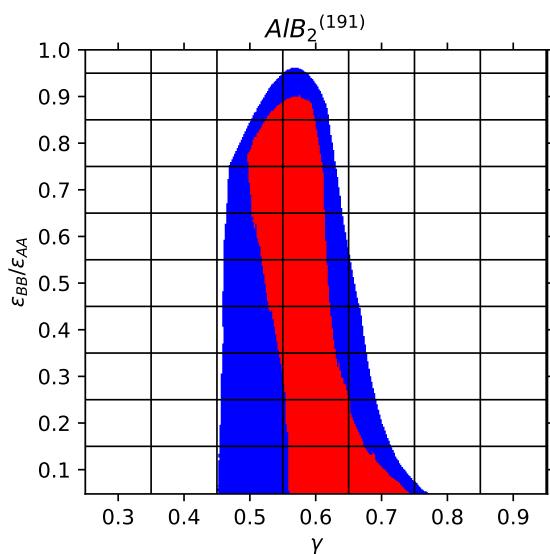
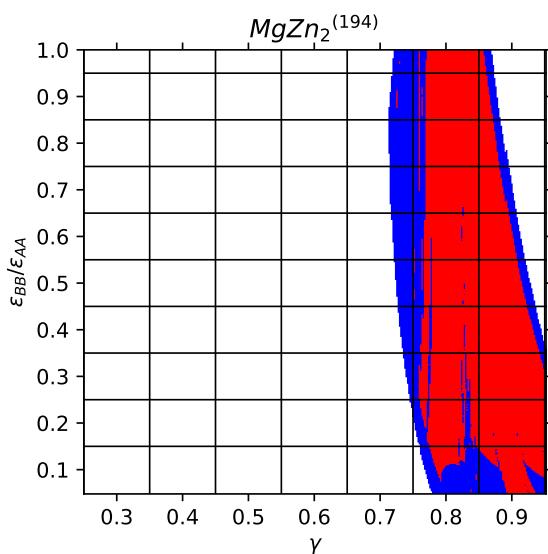
$A_6B_{12}^{(166)}_{(2)}$	0.8	166	Motif-16-2	3.7 6.9 2.1 117.8 30.0 128.9	A(6c) 0.0 0.0 -0.042 A(6c) 0.0 0.0 0.181 A(6c) 0.0 0.0 0.264 B(9e) 0.5 0.0 0.0 B(6c) 0.0 0.0 0.389	
$A_6B_4^{(141)}_{(3)}$	0.5	141	Motif-6-1 Motif-14-1	2.2 2.2 10.0 83.8 83.8 90.0	B(18h) 0.167 0.833 0.111 B(3b) 0.0 0.0 0.5	
$A_2B_{18}^{(15)}_{(2)}$	0.6	15	Motif-22-1	2.7 3.8 2.9 100.0 62.6 112.5	A(8e) 0.0 0.25 0.297 A(4a) 0.0 0.75 0.125 B(8e) 0.0 0.25 -0.016	
$AB_6^{(229)}_{(1)}$	0.5	229	Motif-24-3	2.9 4.1 2.5 90.0 54.7 135.0	A(4e) 0.0 0.326 0.25 B(4e) 0.0 -0.06 0.25 B(8f) -0.068 0.247 0.605 B(8f) 0.463 0.408 0.546 B(8f) 0.321 0.053 0.019 B(8f) 0.806 0.41 0.795	
$A_2B_{18}^{(63)}_{(4)}$	0.5	63	Motif-27-1	2.9 3.7 3.7 90.0 50.4 90.0	A(2a) 0.0 0.0 0.0 B(12d) 0.25 0.0 0.5 A(4c) 0.0 0.775 0.25 B(4c) 0.0 0.226 0.25 B(16h) 0.687 0.386 0.896 B(16h) 0.848 0.4 0.109	
$A_2B_{10}^{(12)}_{(14)}$	0.5	12	Motif-18-4	4.1 2.1 3.0 111.0 102.6 75.4	A(4i) 0.281 0.0 0.316 B(4g) 0.0 0.254 0.0 B(4i) 0.024 0.0 0.386 B(4i) 0.53 0.0 0.237 B(4i) 0.33 0.0 -0.0 B(4i) 0.104 0.0 0.886	

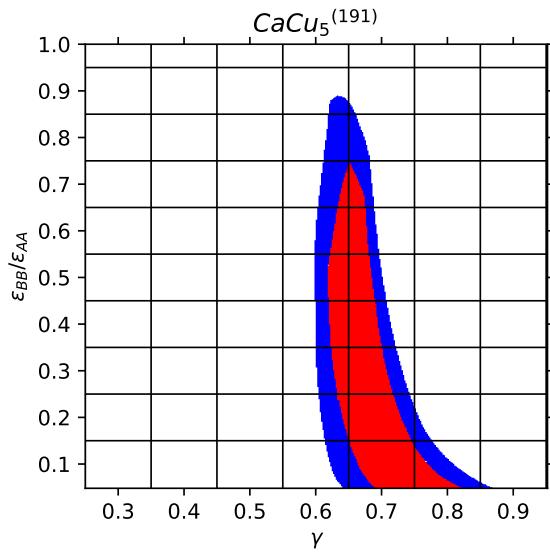
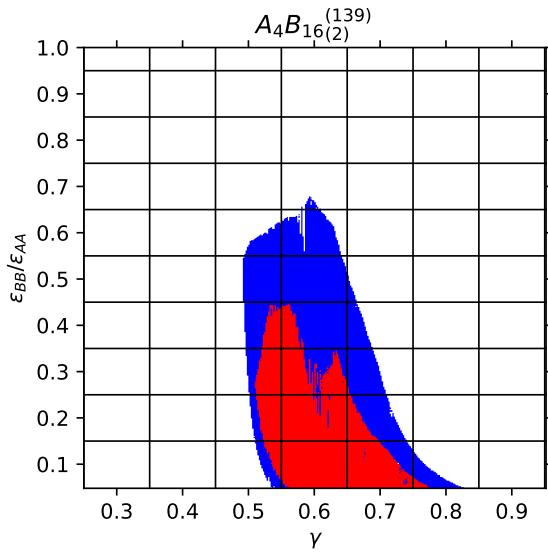
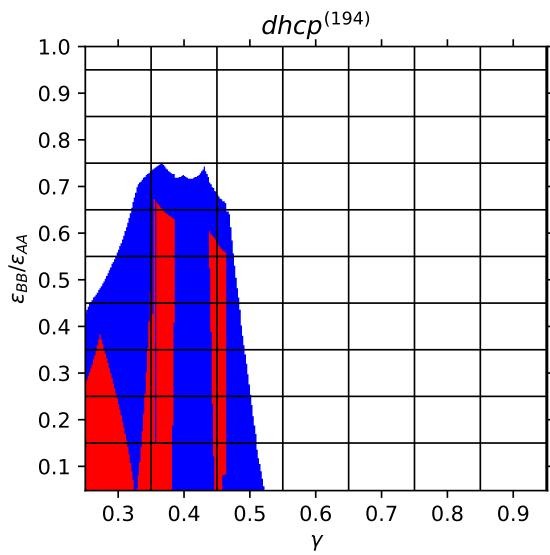
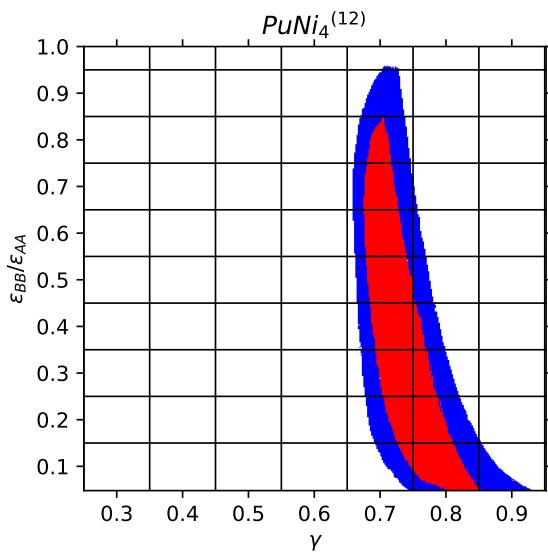
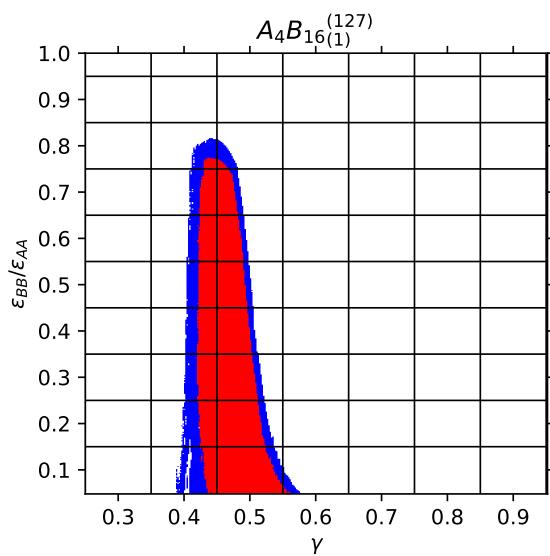
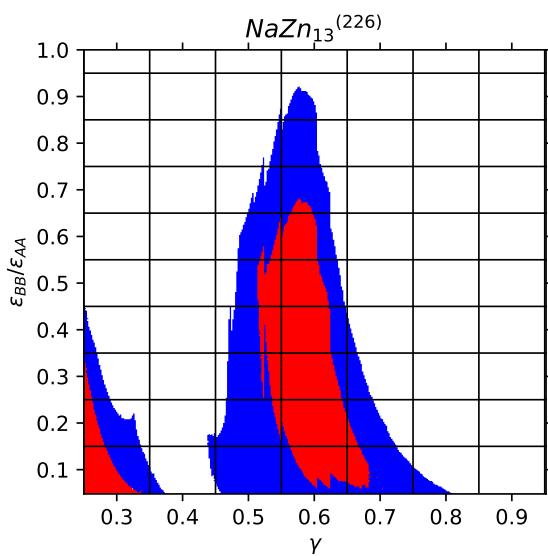
$A_2B_{18}^{(63)}_{(3)}$	0.4	63	Motif-29-1	3.6 8.3 2.6 90.0 68.7 139.7	A(4c) 0.0 0.088 0.25 B(8f) 0.0 0.714 0.478 B(8f) 0.0 0.431 0.047 B(16h) 0.2 0.17 0.875 B(4c) 0.0 0.378 0.25
$A_4B_2^{(141)}_{(8)}$	0.4	141	Motif-3-2	4.7 4.7 4.7 131.7 70.7 131.7	A(8e) 0.0 0.25 0.626 B(4b) 0.0 0.25 0.375
$A_2B_{18}^{(15)}_{(3)}$	0.4	15	Motif-29-2	2.6 4.8 3.7 90.9 69.0 92.6	A(4e) 0.0 0.085 0.25 B(8f) 0.78 0.177 0.894 B(4e) 0.0 0.377 0.25 B(8f) 0.486 0.07 -0.046 B(8f) 0.169 0.176 0.841 B(8f) 0.05 0.284 -0.015
$A_5B_{10}^{(12)}_{(2)}$	0.3	12	Motif-6-4 Motif-15-2	3.1 3.1 13.0 100.1 83.3 119.7	A(4i) 0.634 0.0 0.804 A(4i) 0.269 0.0 0.608 A(2a) 0.0 0.0 0.0 B(4h) 0.0 0.75 0.5 B(4i) -0.068 0.0 0.529 B(4i) 0.317 0.0 -0.098 B(4i) 0.422 0.0 0.471 B(4i) 0.049 0.0 0.294
$AB_6^{(65)}_{(1)}$	0.6	65	Motif-18-2	2.7 5.4 4.2 141.1 18.5 149.4	A(2a) 0.0 0.0 0.0 B(4j) 0.0 0.397 0.5 B(4i) 0.0 0.708 0.0 B(4j) 0.0 0.202 0.5
$A_2B_{12}^{(148)}_{(1)}$	0.5	148	Motif-21-2	3.0 3.7 3.7 120.0 52.2 127.8	A(6c) 0.0 0.0 0.175 B(18f) 0.171 0.679 0.236 B(18f) 0.651 -0.046 0.018

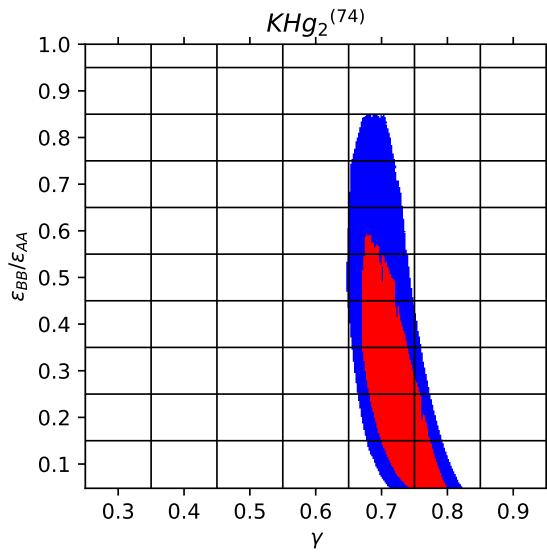
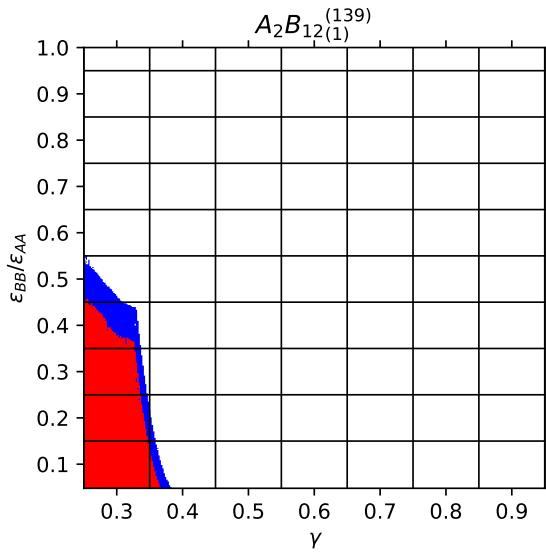
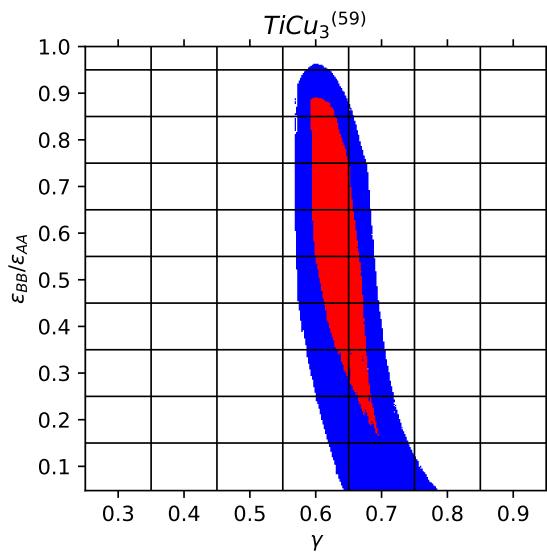
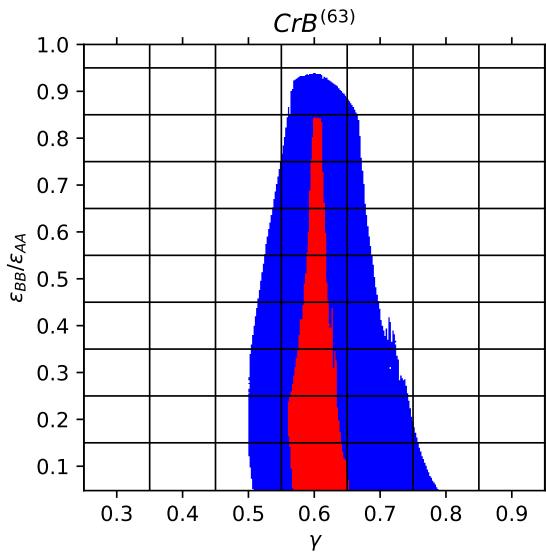
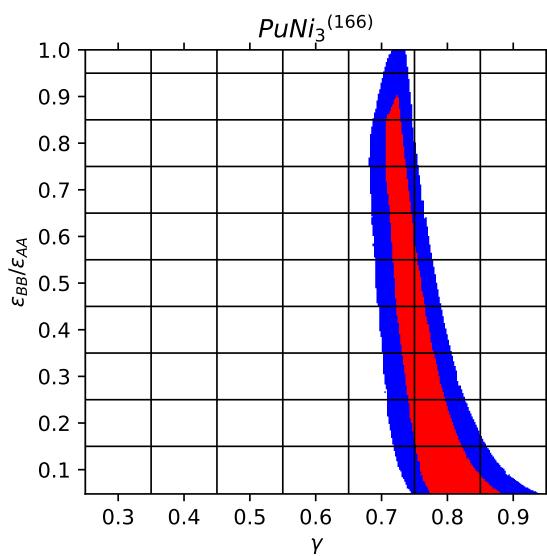
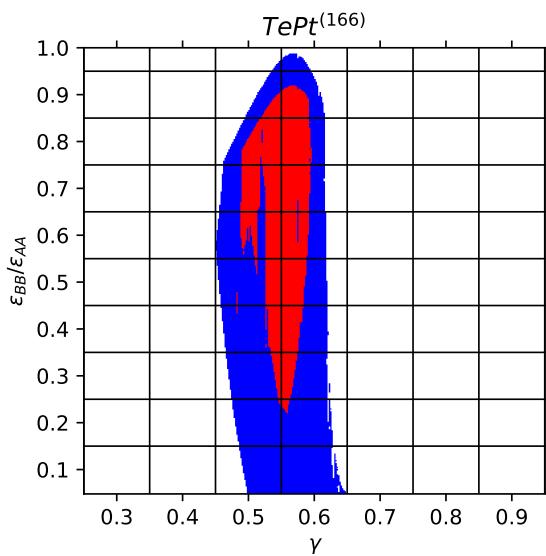
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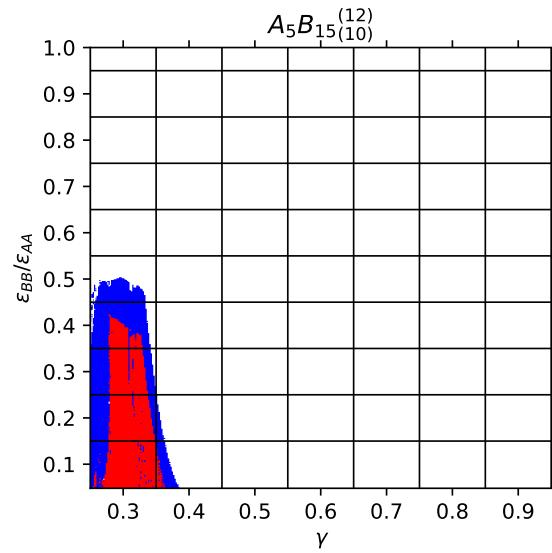
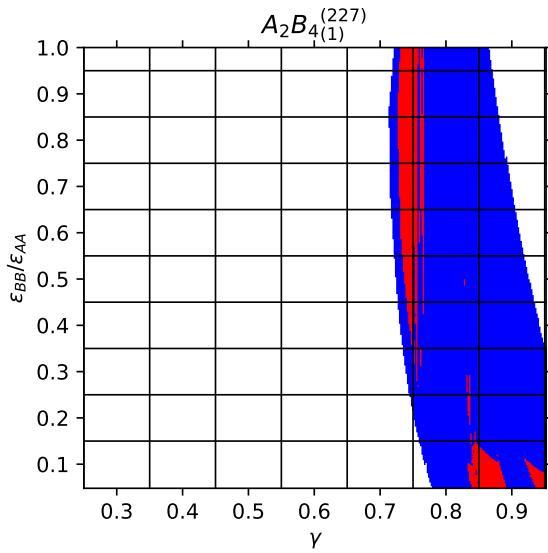
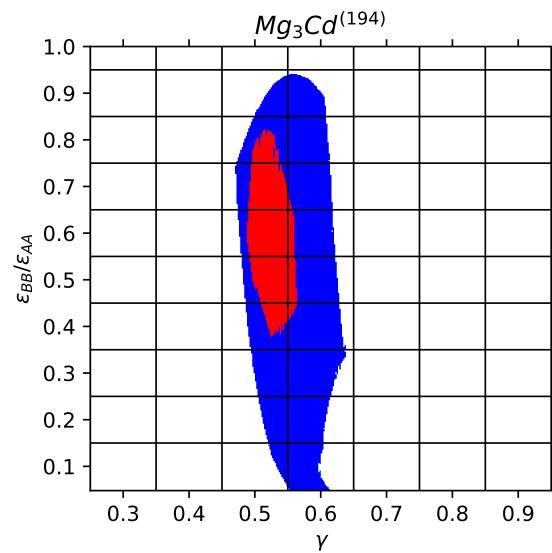
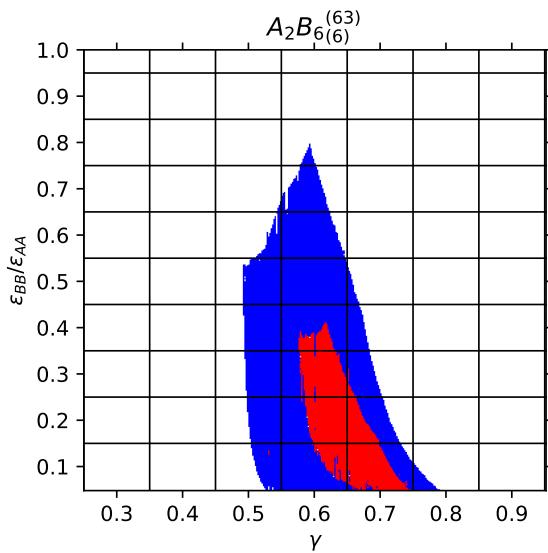
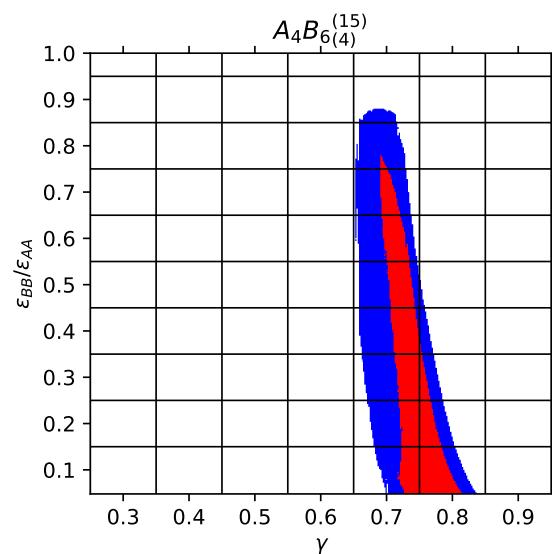
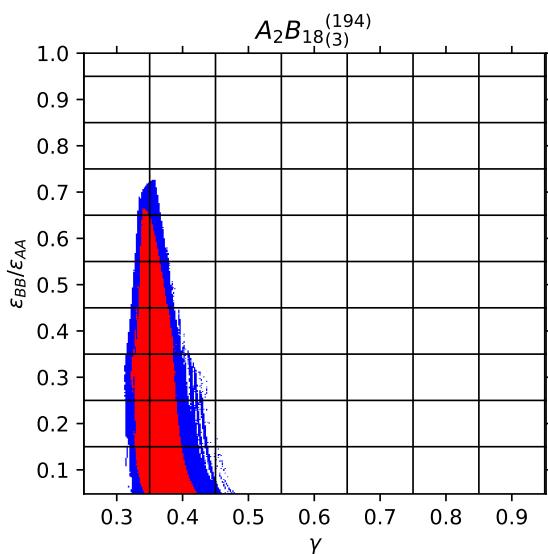
$A_2B_{12}^{(12)}_{(9)}$	0.3	12	Motif-21-1	2.9 5.5 4.2 95.4 45.3 105.5	A(4i) 0.863 0.0 0.807 B(4i) 0.465 0.0 0.576 B(4i) 0.413 0.0 0.297 B(8j) 0.027 0.268 0.642 B(4i) 0.516 0.0 0.86 B(4i) 0.326 0.0 0.297
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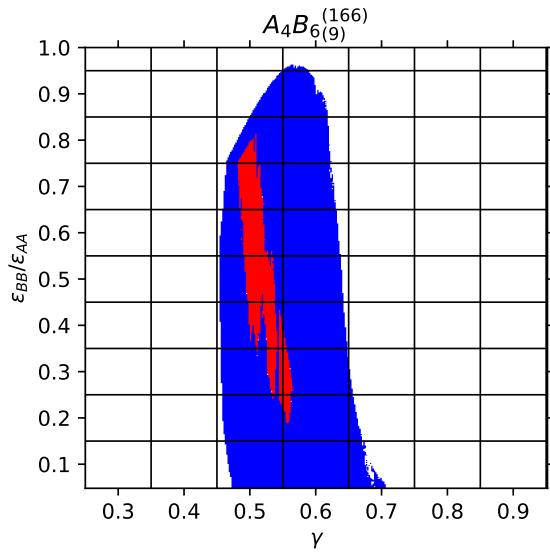
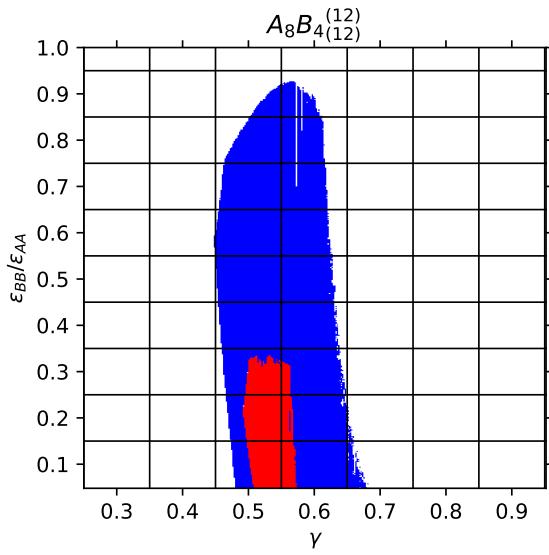
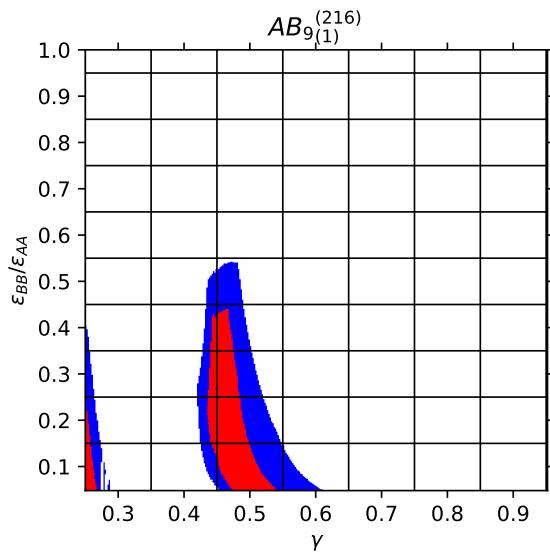
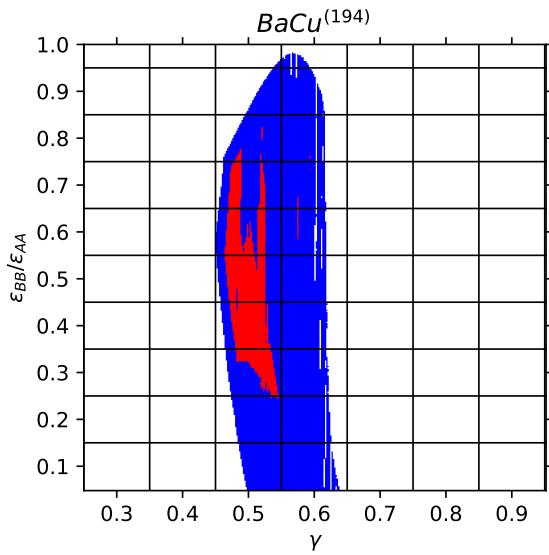
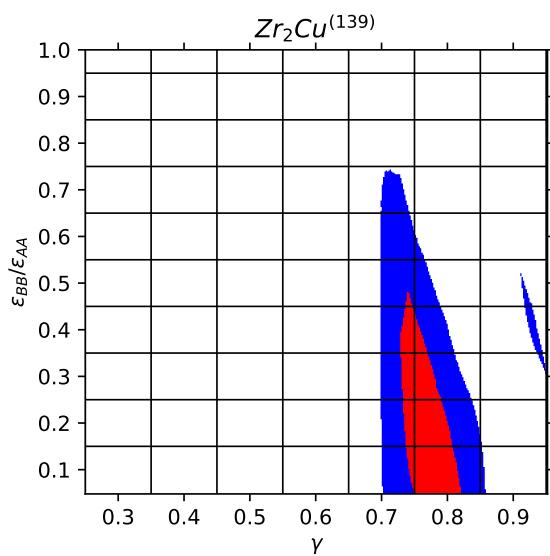
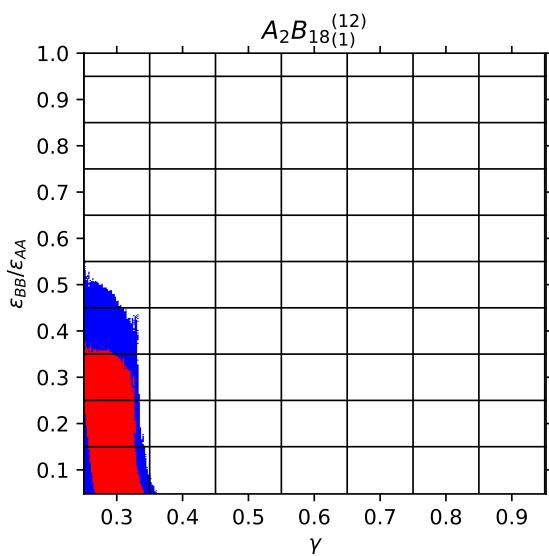
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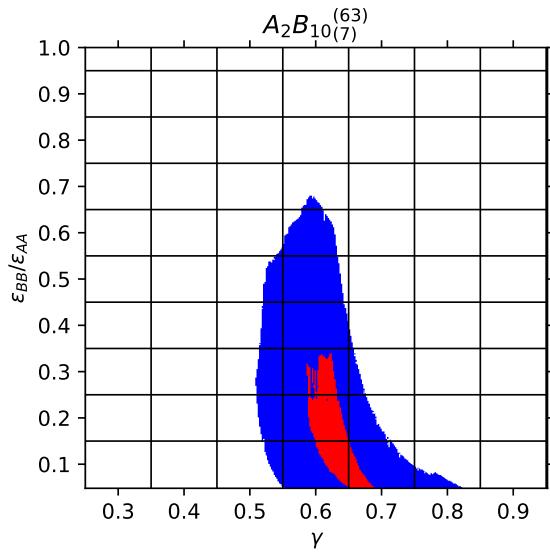
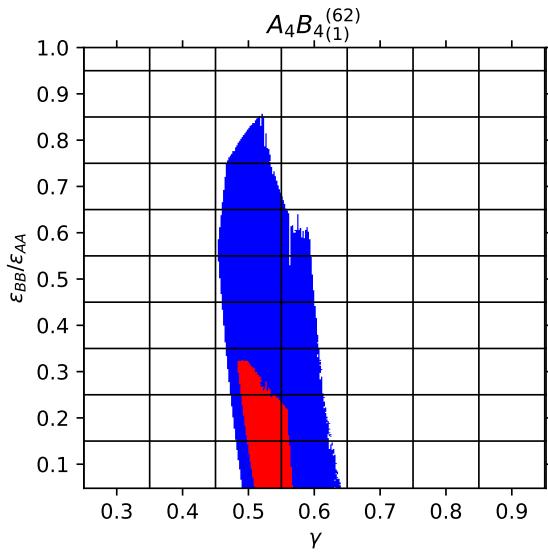
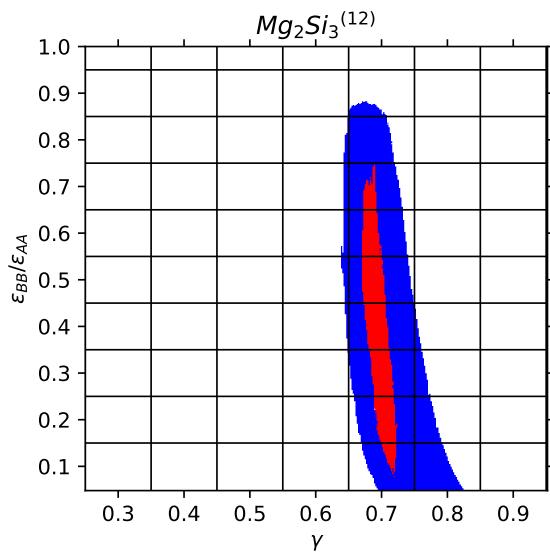
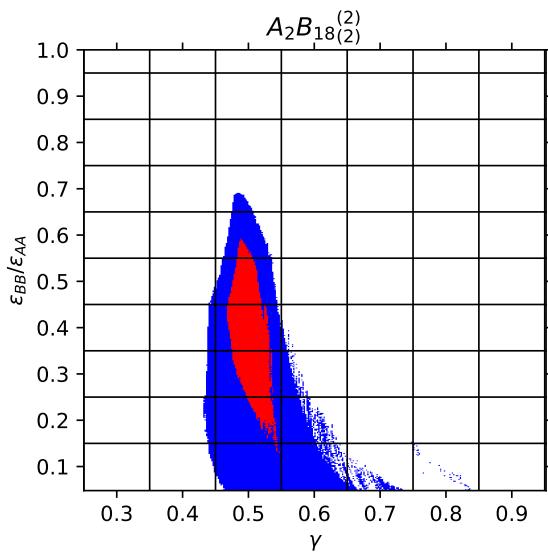
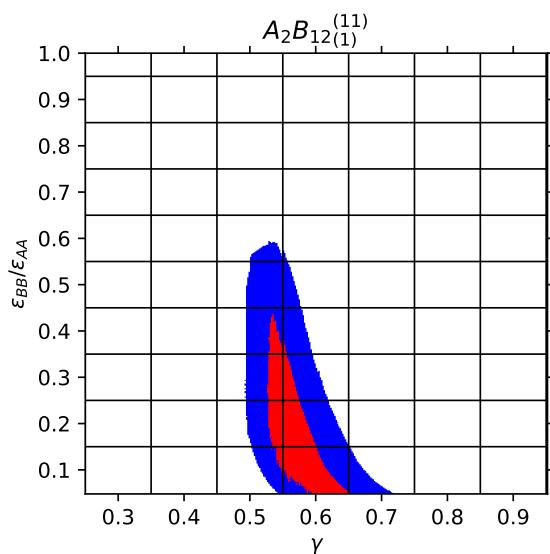
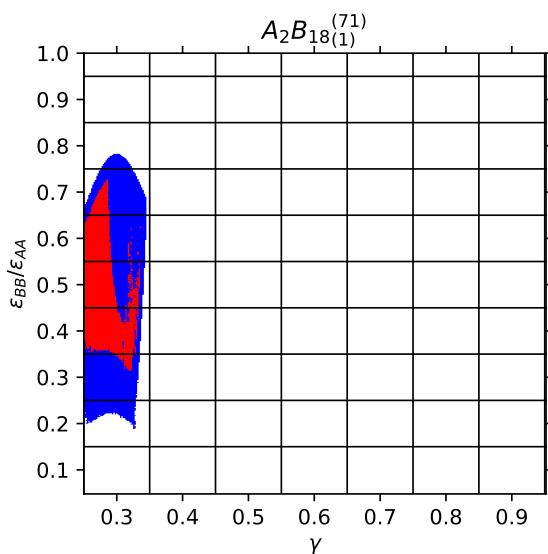


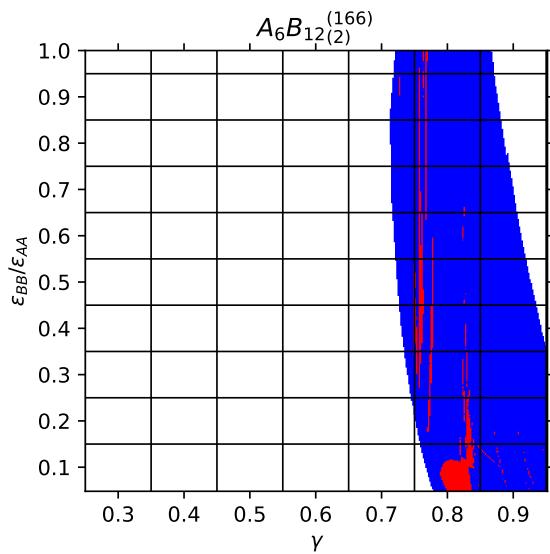
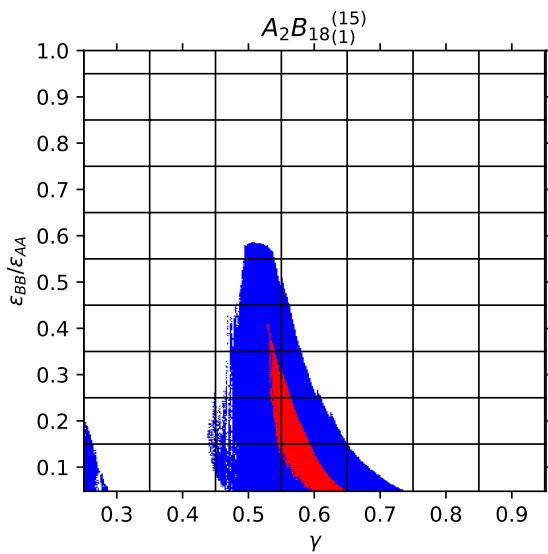
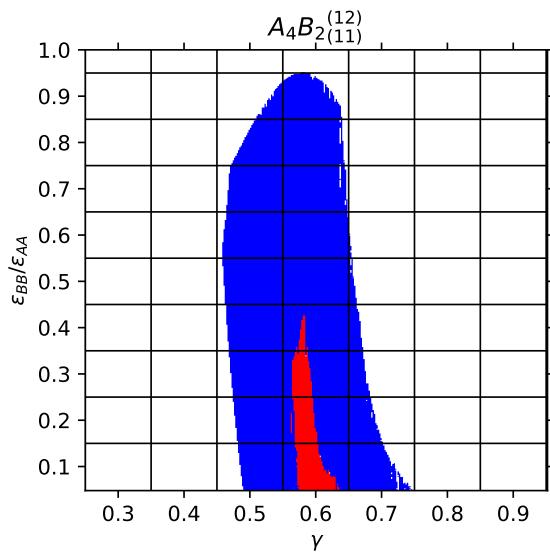
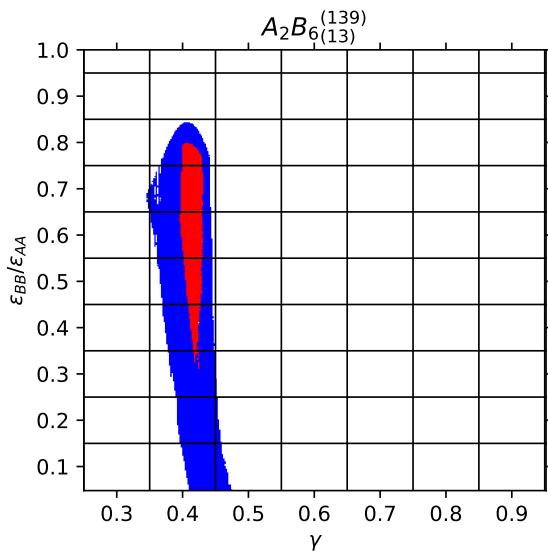
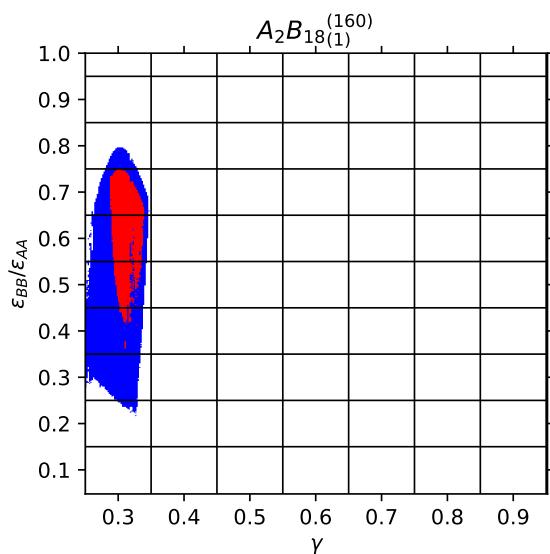
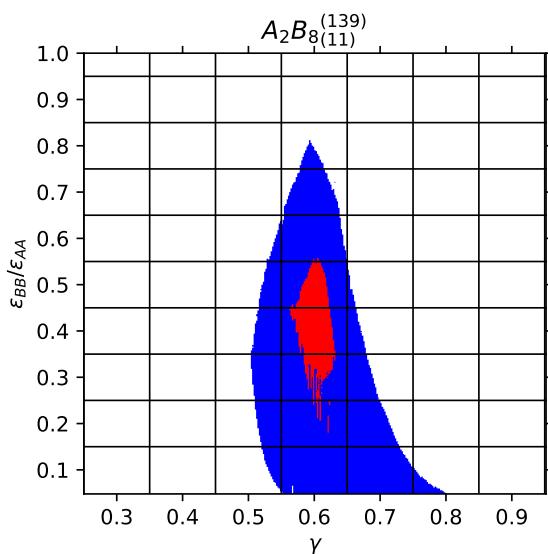


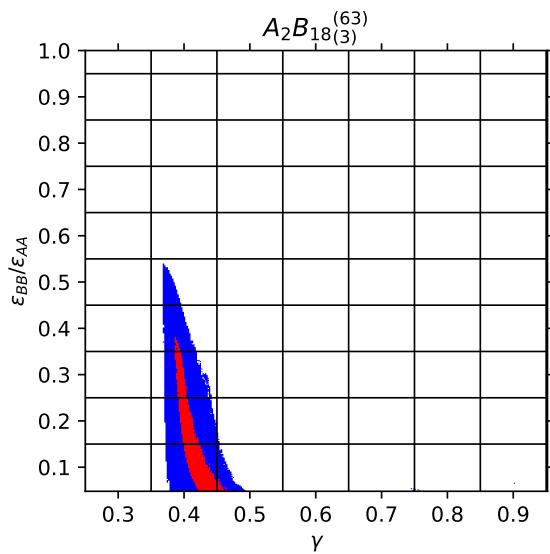
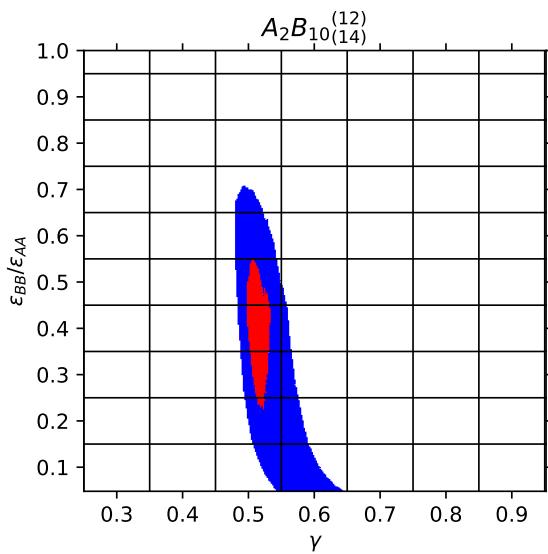
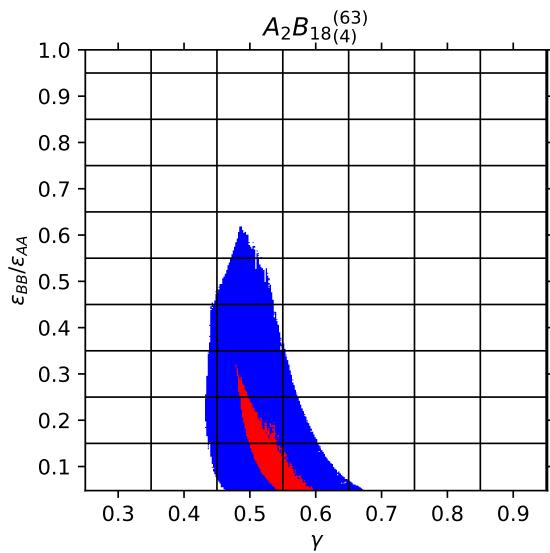
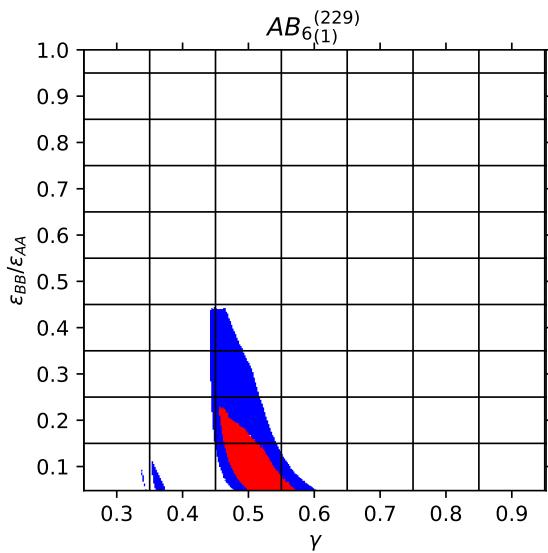
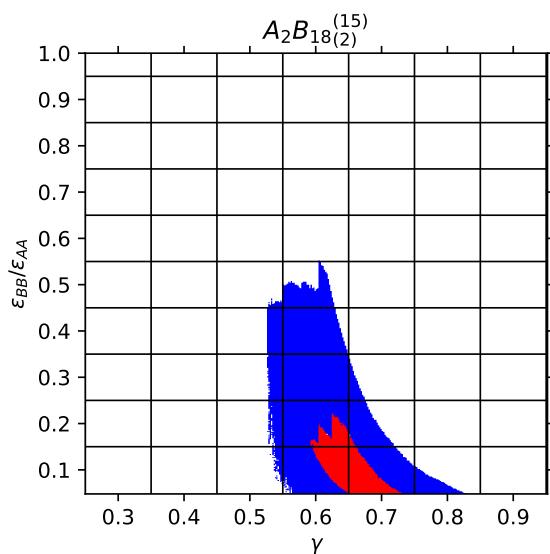
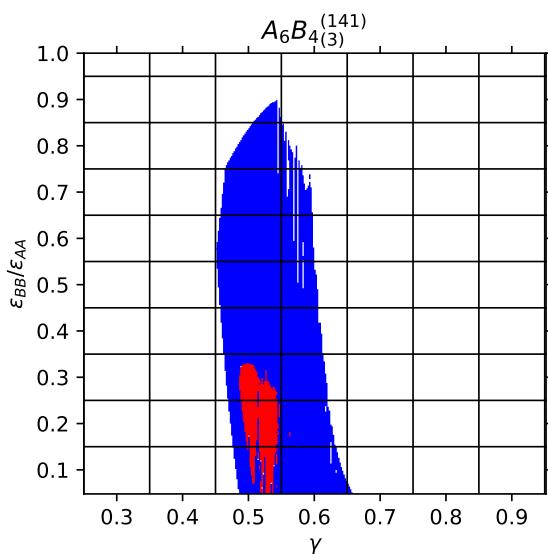


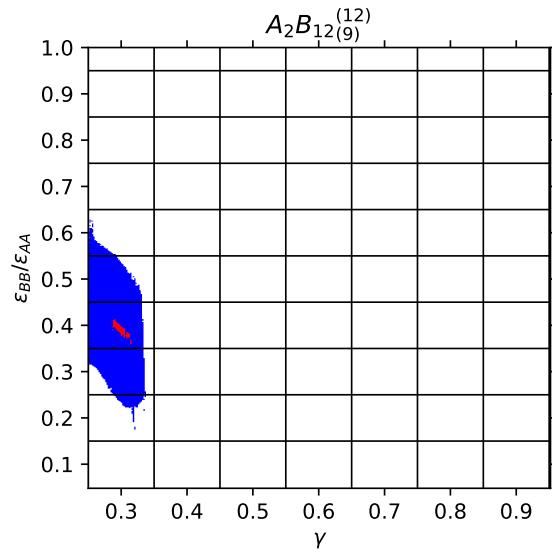
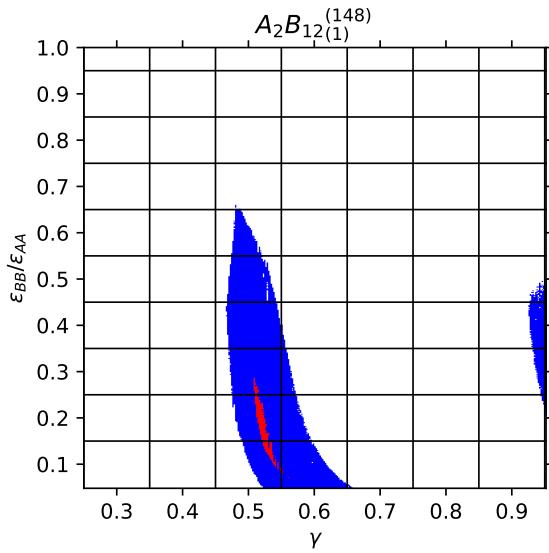
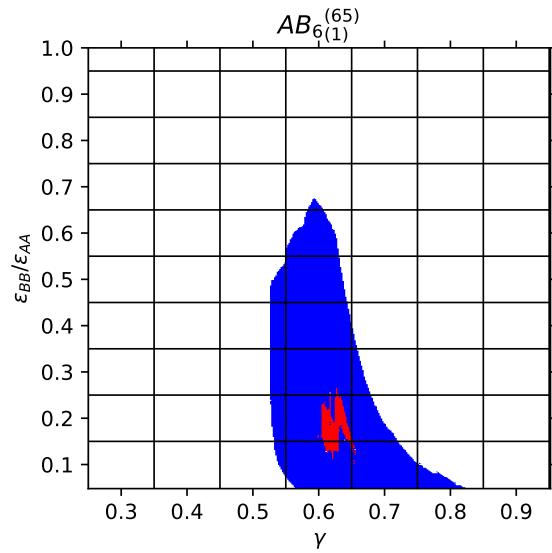
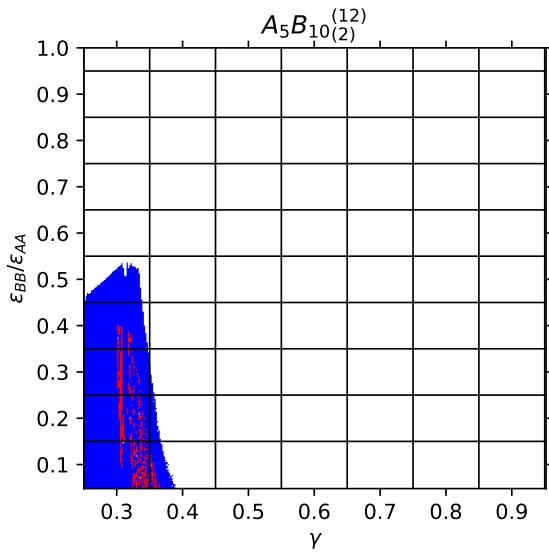
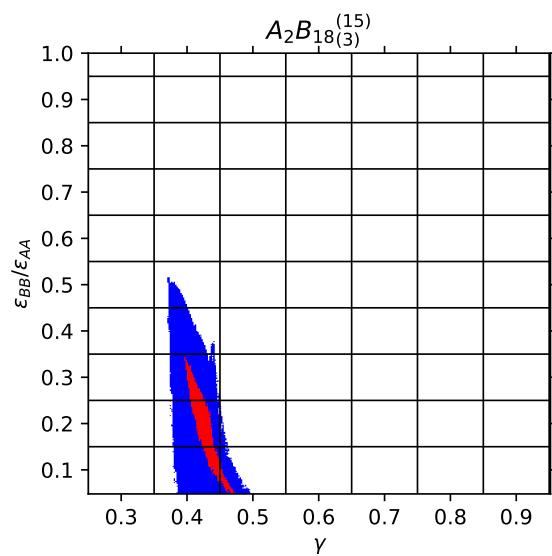
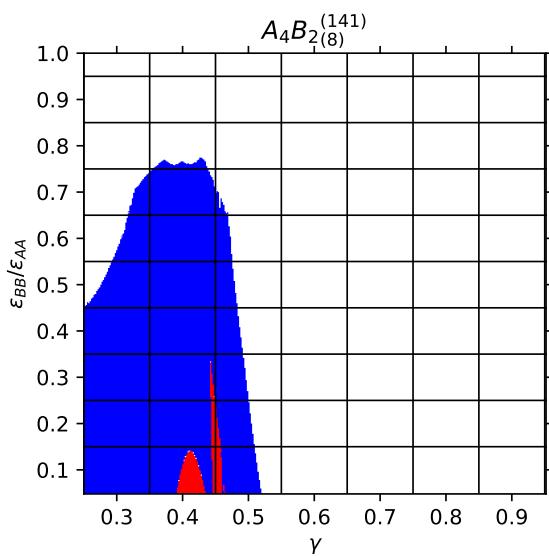












## Phase diagrams of equilibrium structures

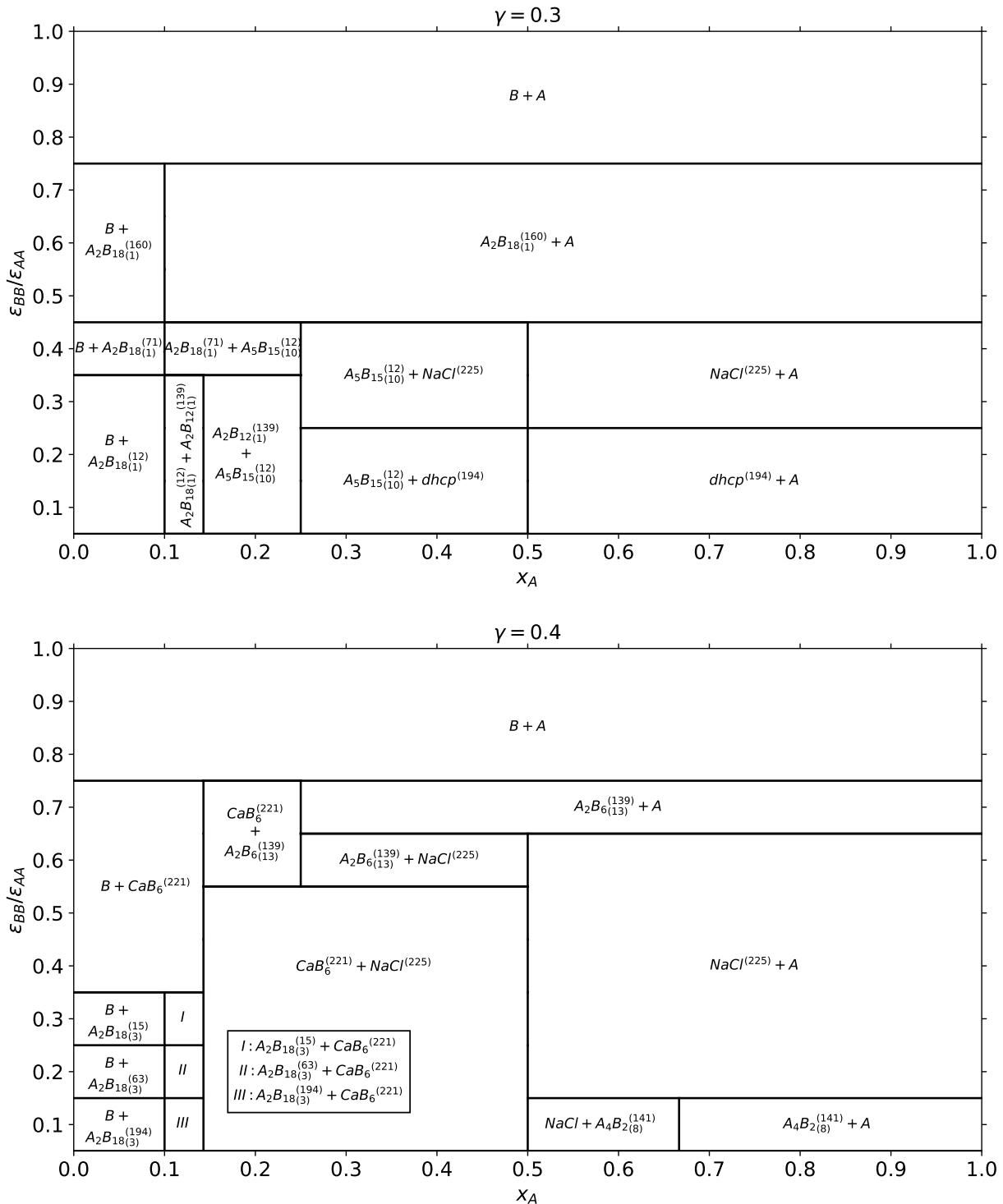


Figure S2

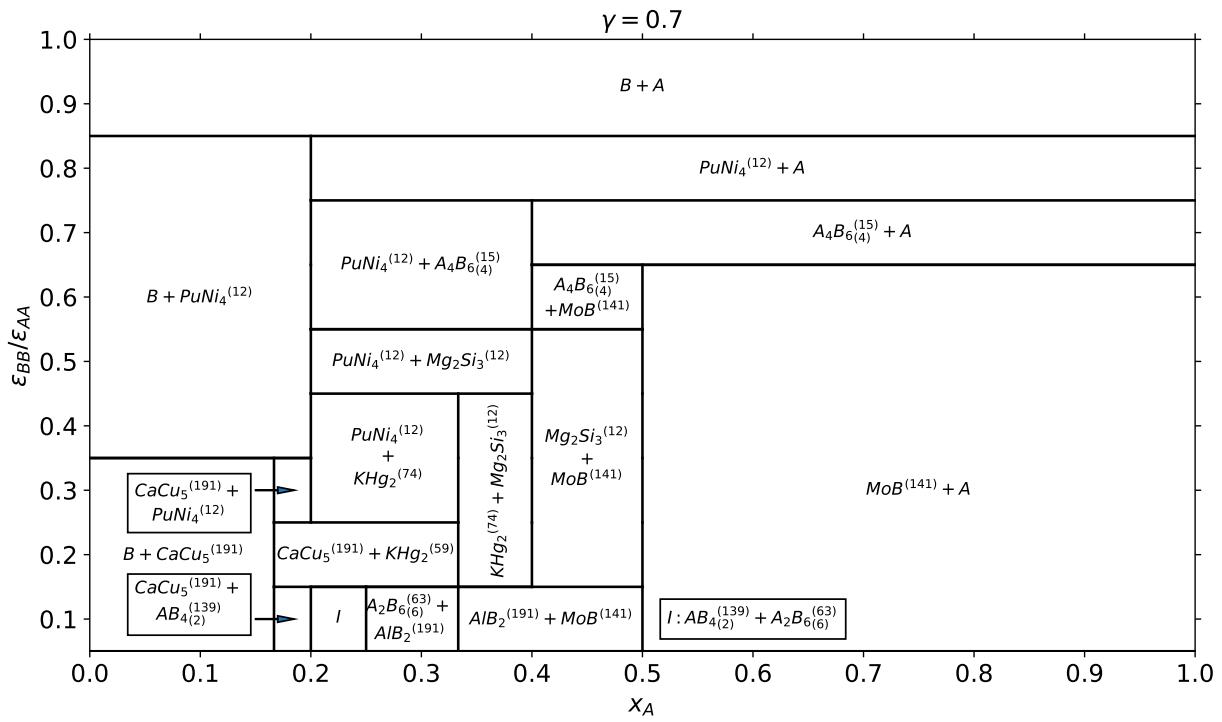
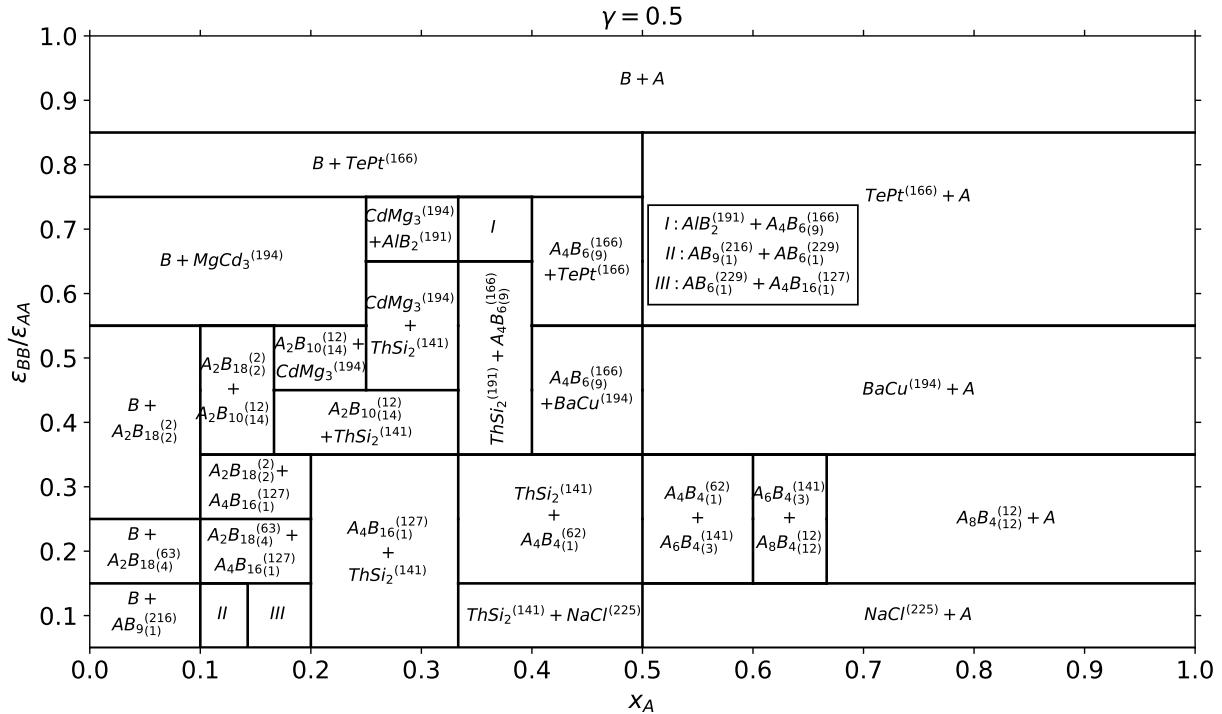


Figure S2

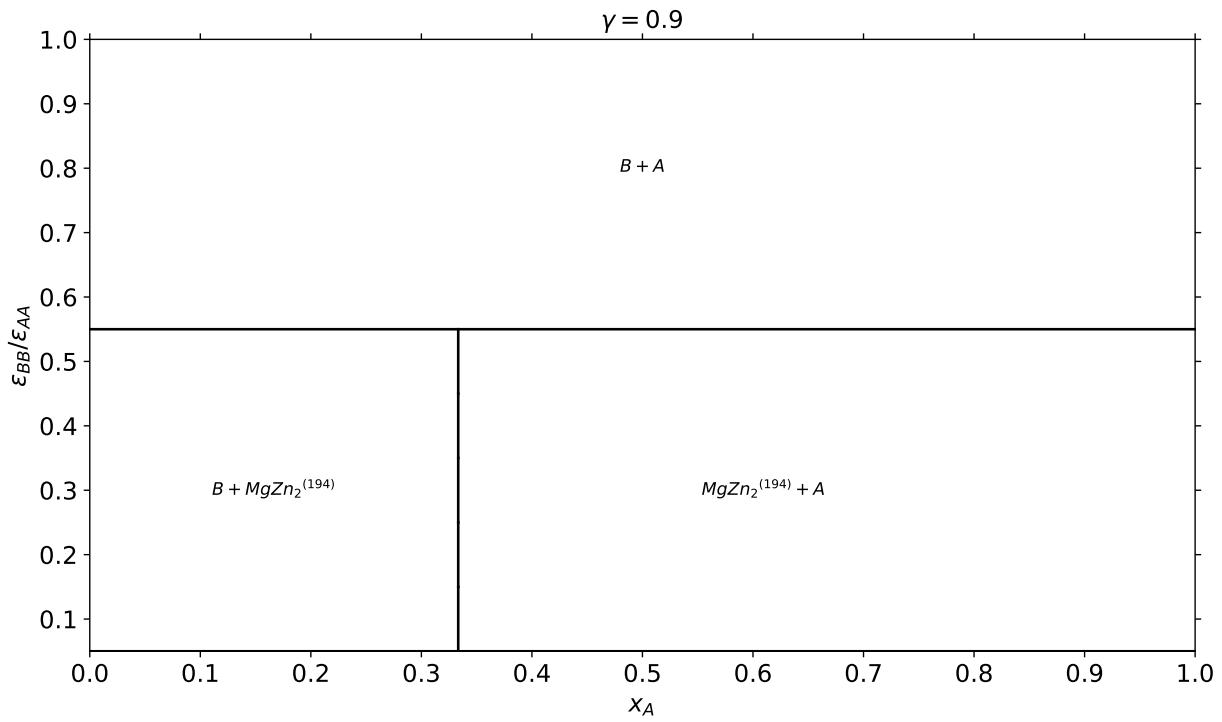
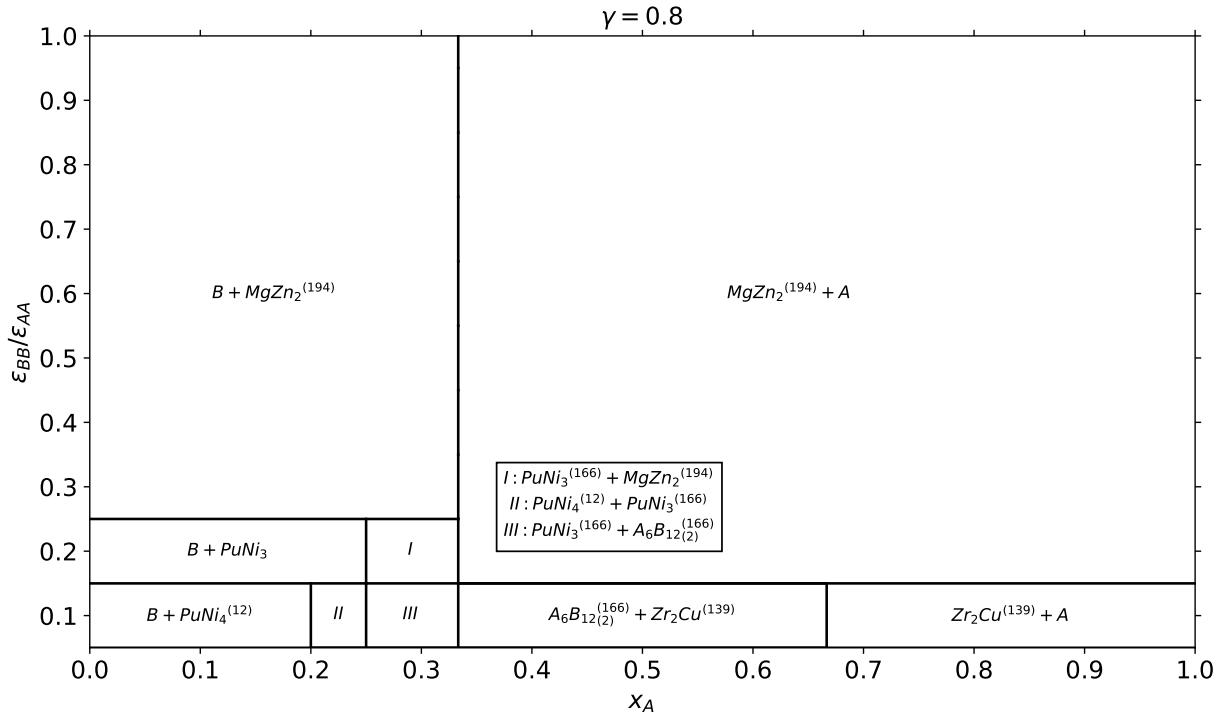


Figure S2

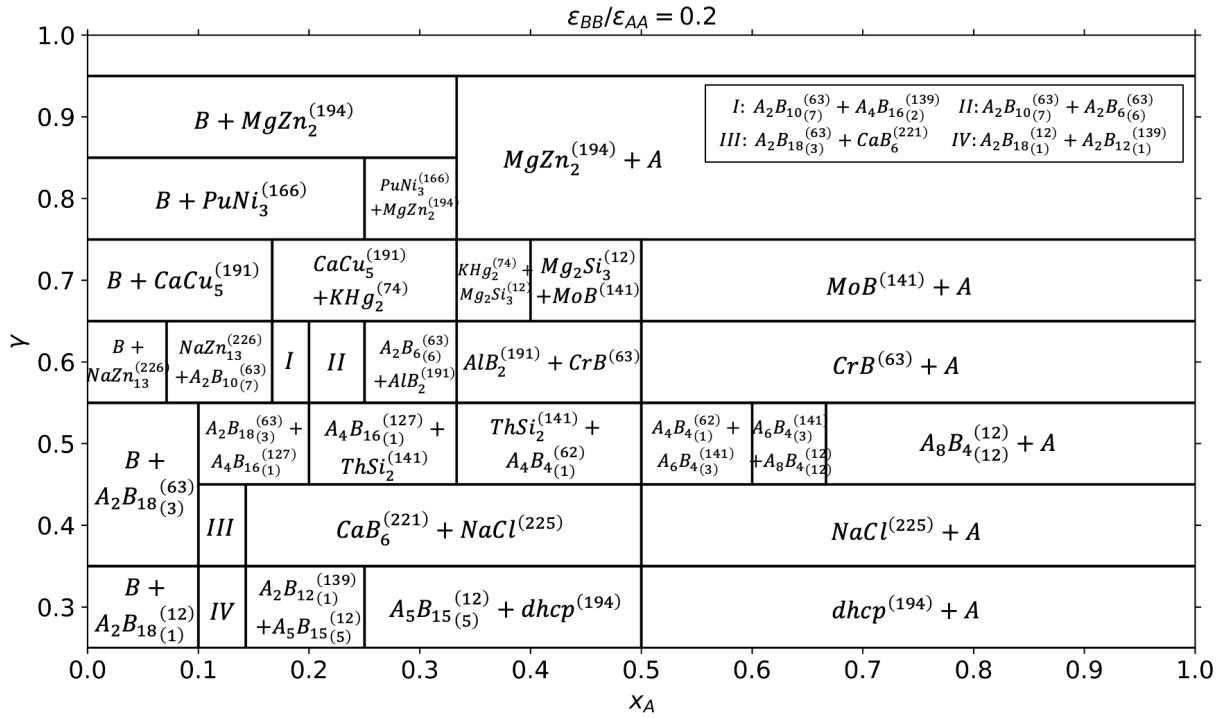
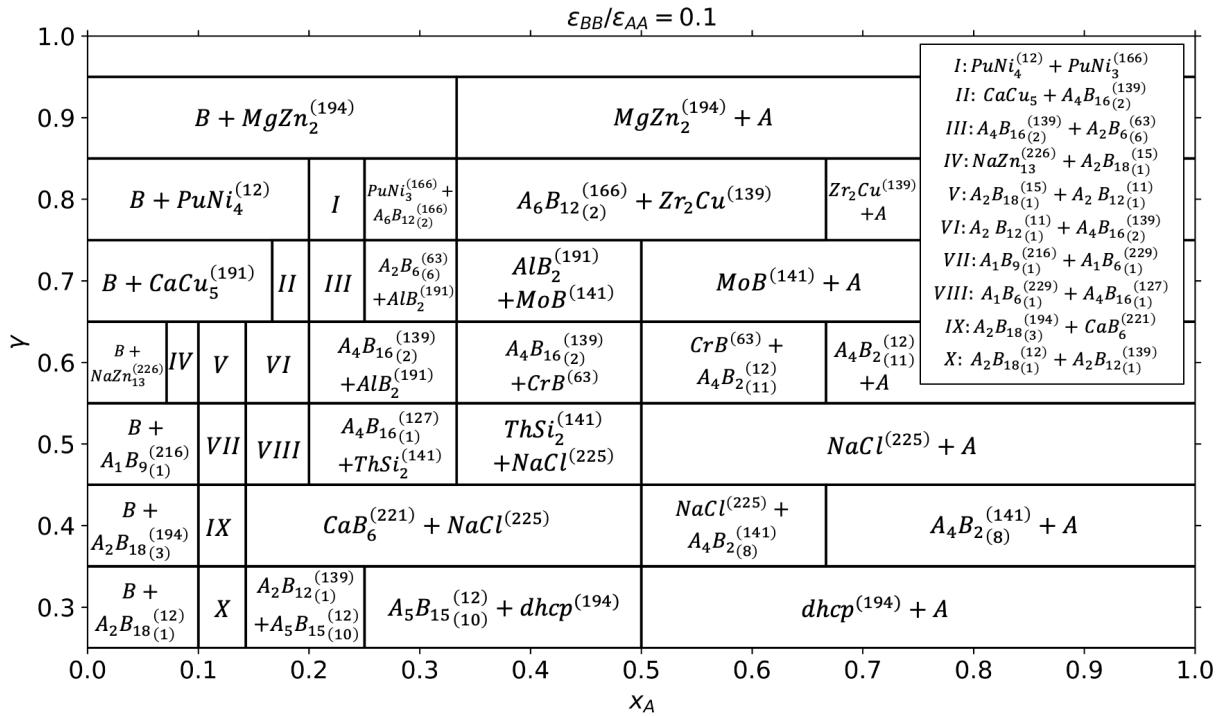


Figure S3

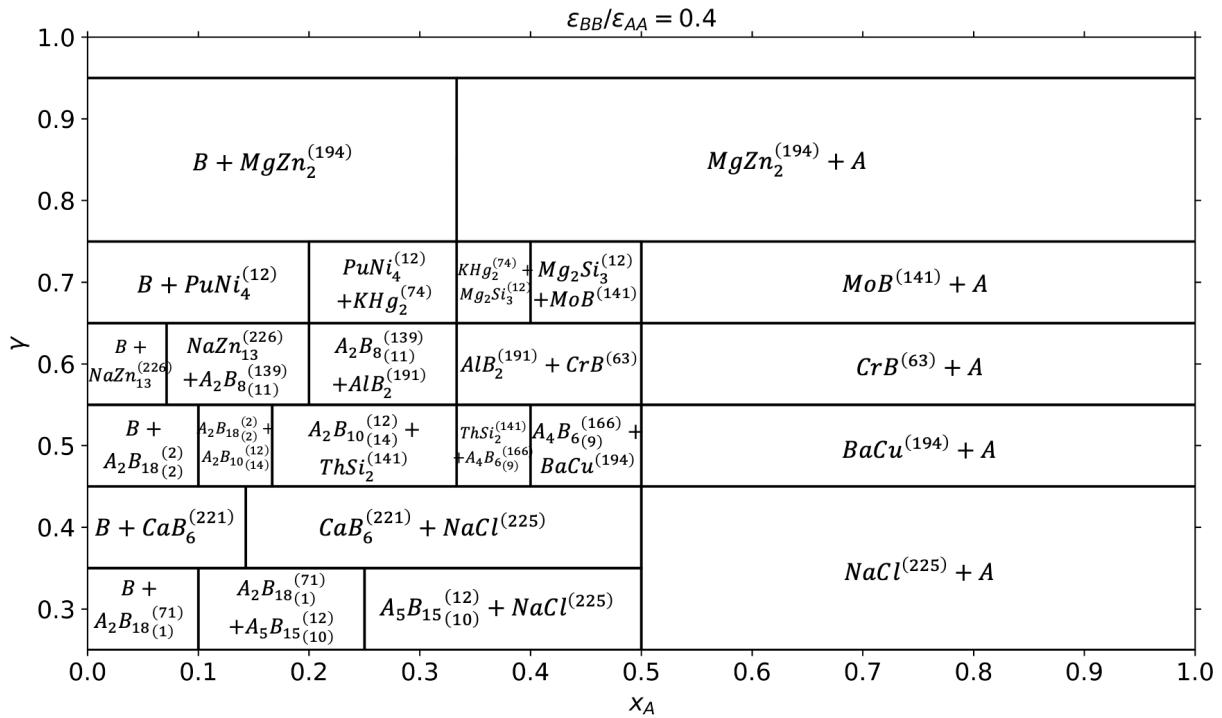
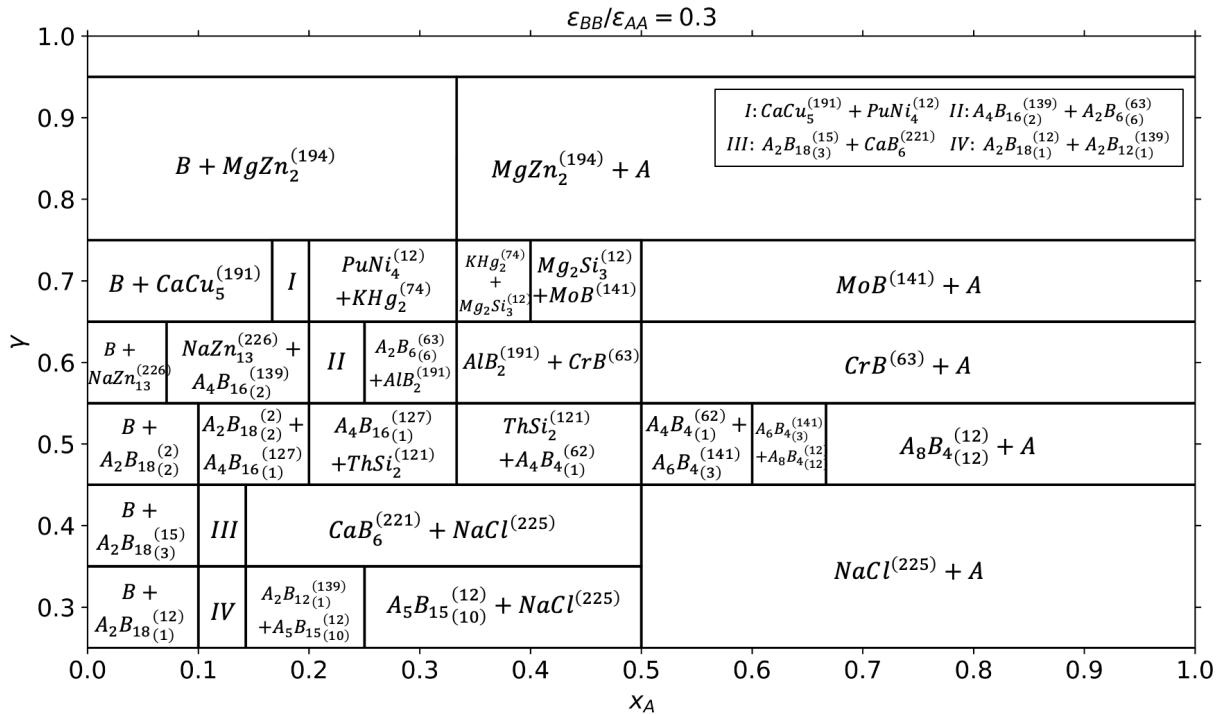


Figure S3

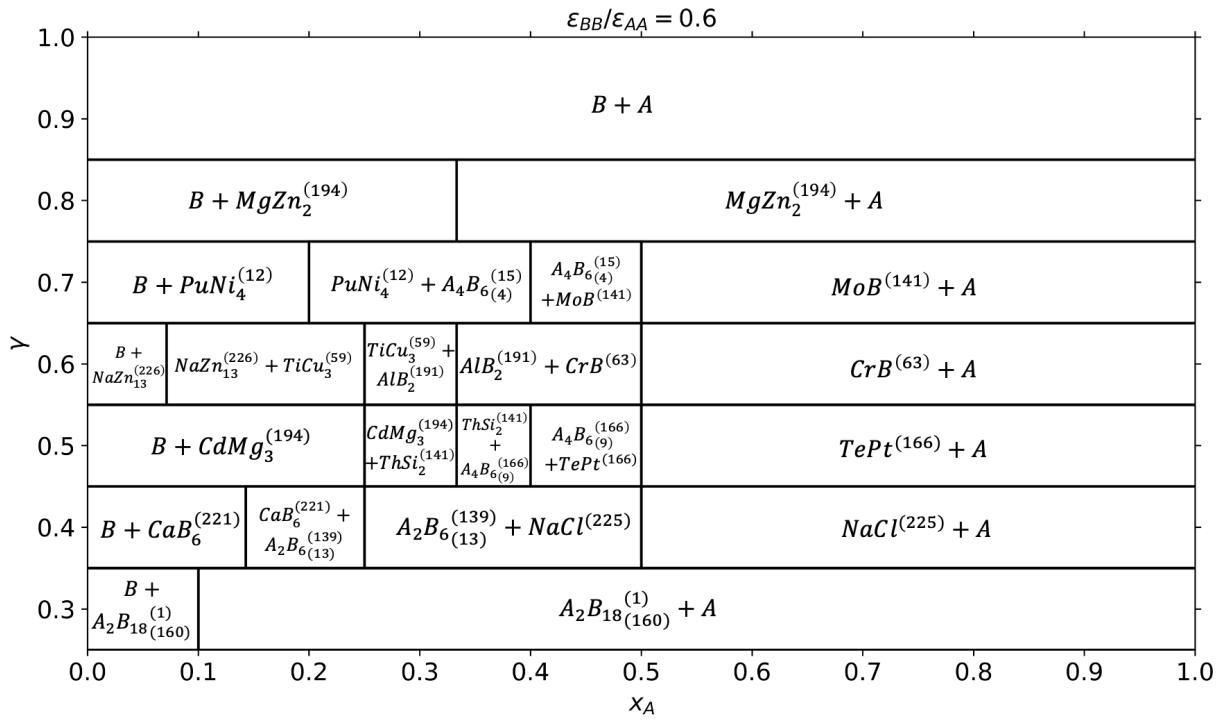
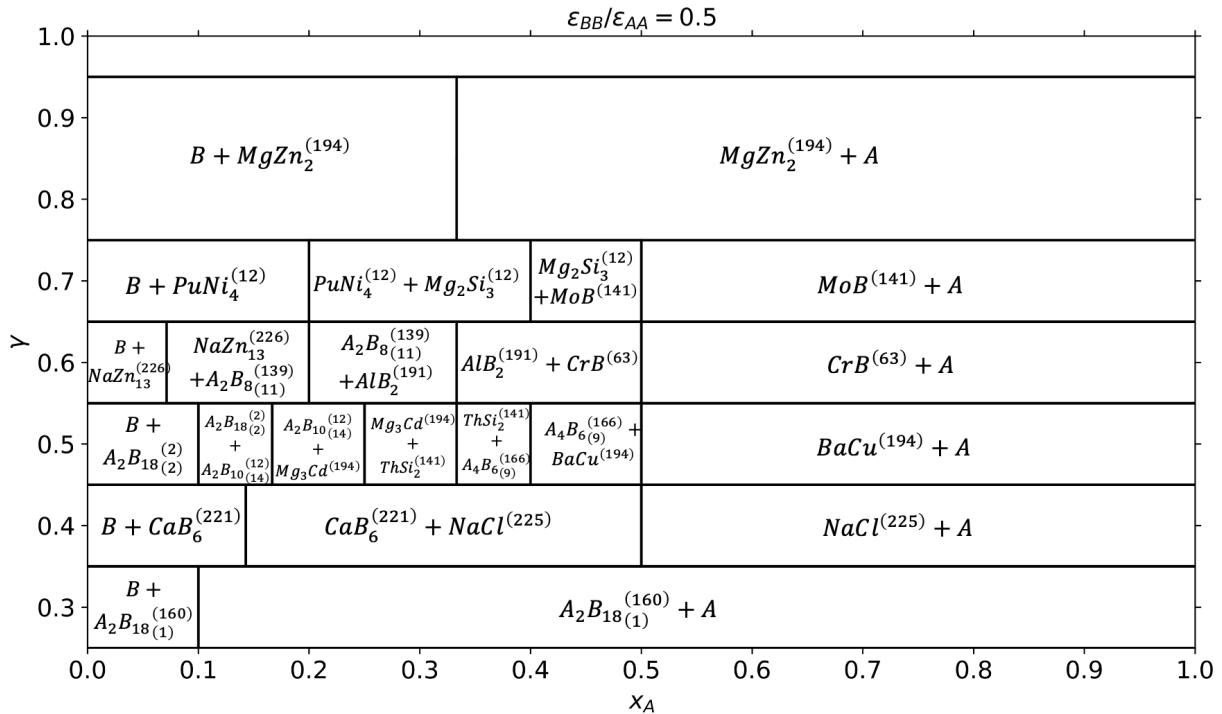


Figure S3

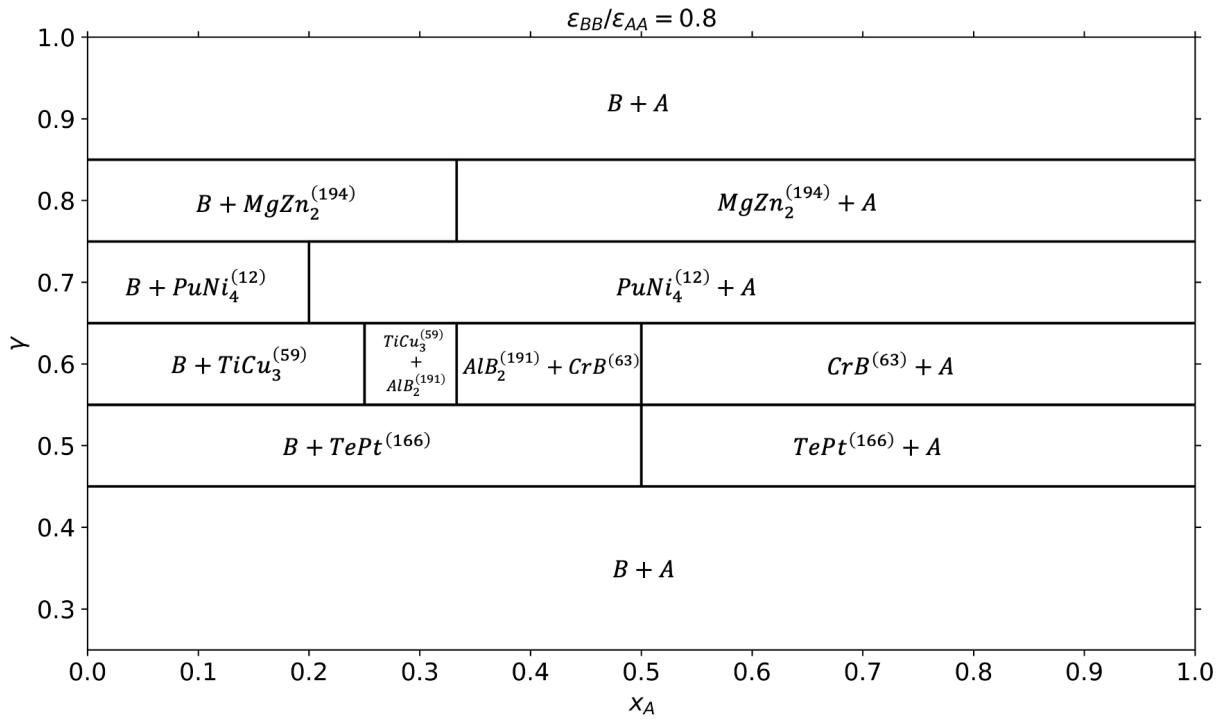
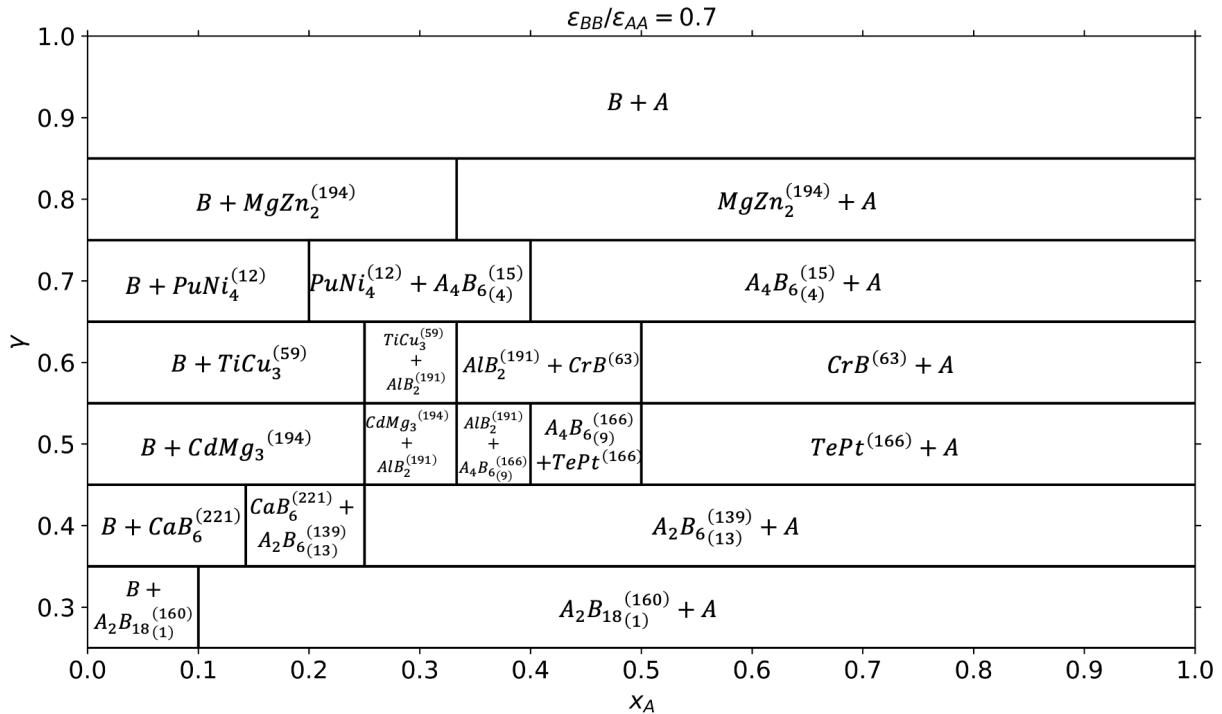


Figure S3

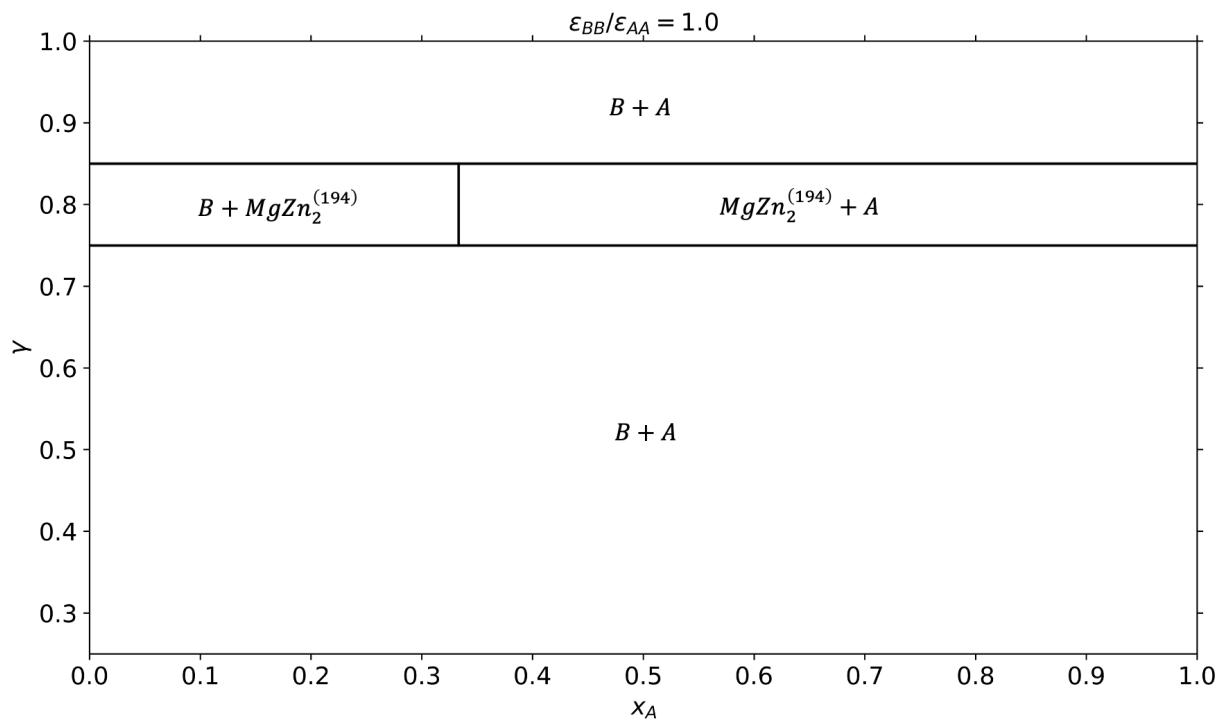
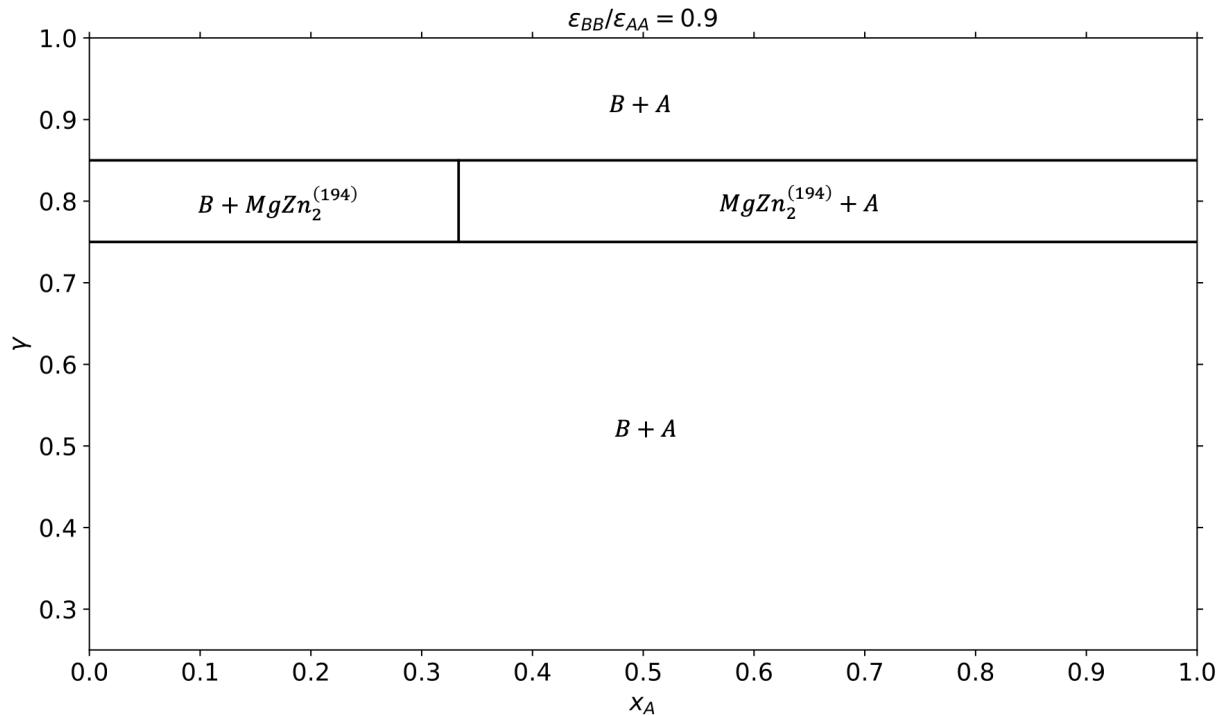
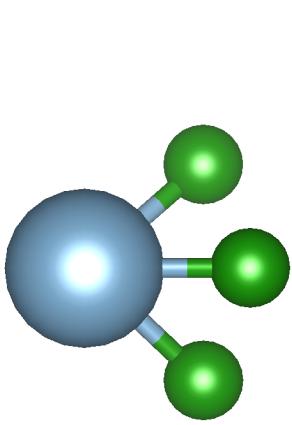


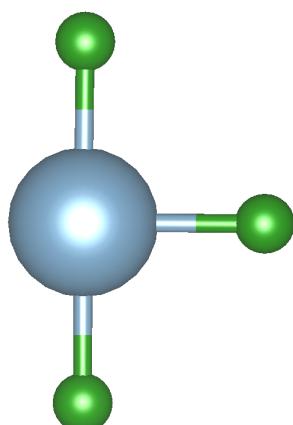
Figure S3

## Equilibrium motif database

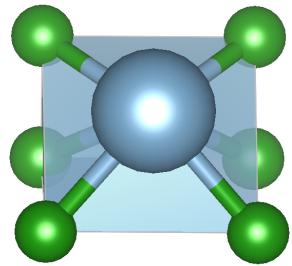
This section contains motifs from equilibrium structures. Each motif (except Motif-3-1 and Motif-3-2) are plotted in polyhedron. Blue particles are A (larger) particles, and green ones are B (smaller) particles. The sizes of particles are assigned by the  $\gamma$  value of each structure. Particles which are neither connected by a bond nor contained in a polyhedron is drawn to show how it affect the motif.



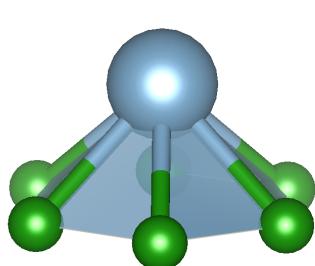
(a) Motif-3-1



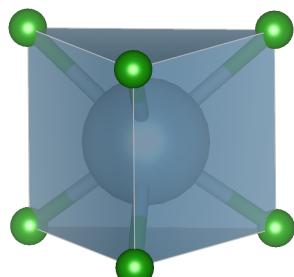
(b) Motif-3-2



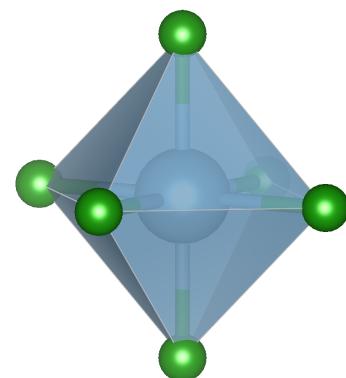
(c) Motif-6-1



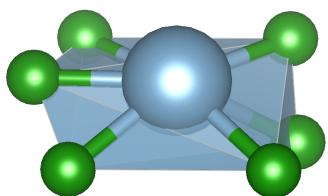
(d) Motif-6-2



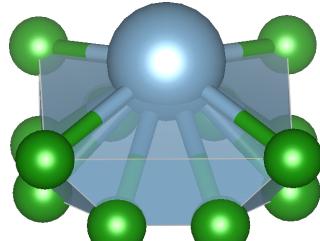
(e) Motif-6-3



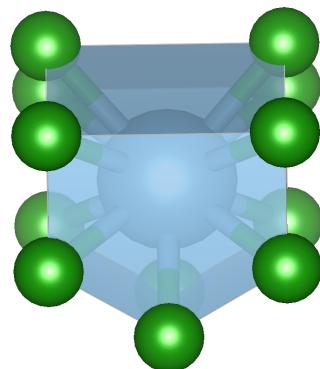
(f) Motif-6-4



(g) Motif-6-5



(h) Motif-12-1



(i) Motif-12-2

Figure S4: Motifs3-1 to Motifs12-2

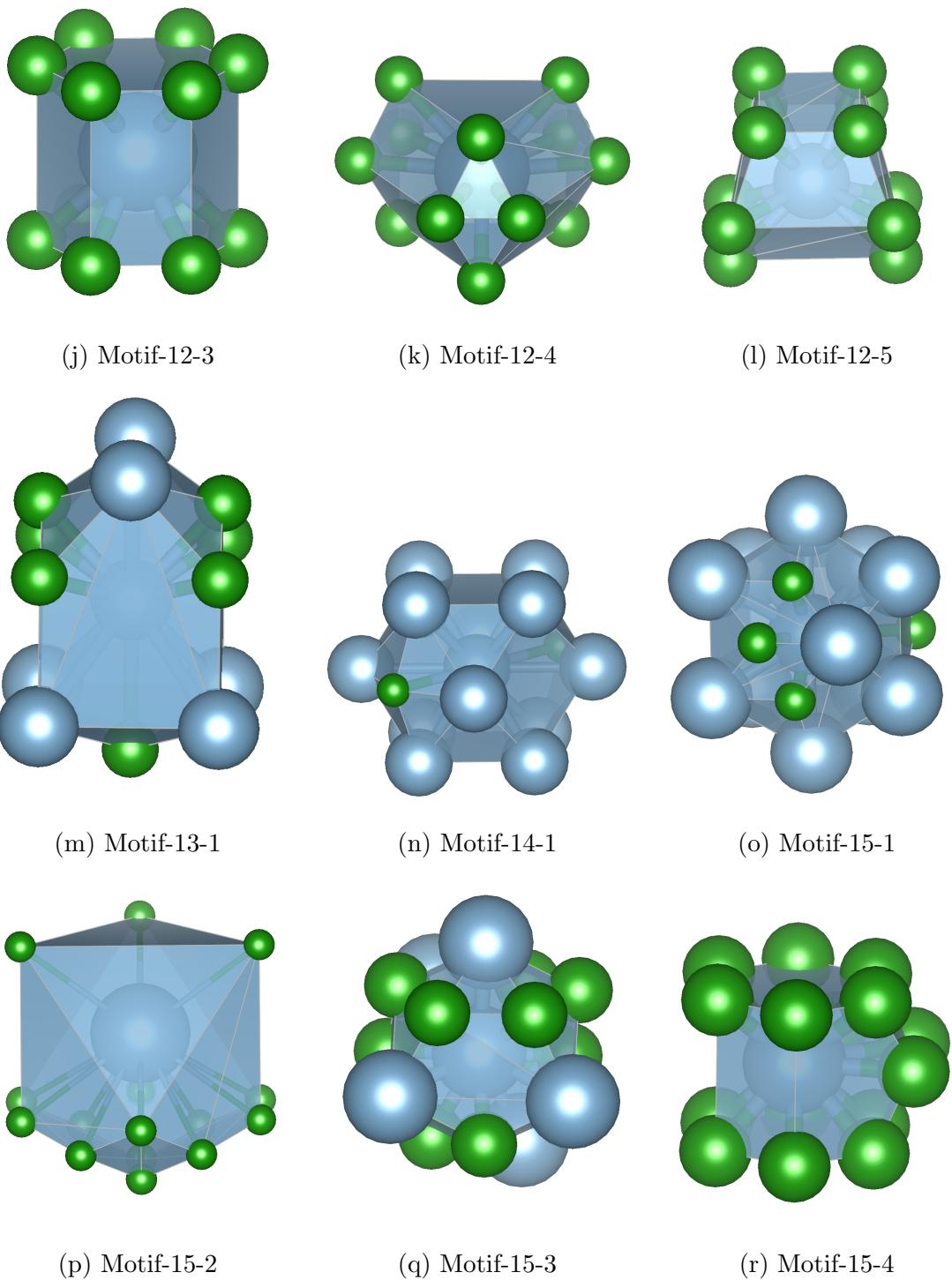


Figure S4: Motifs 12-3 to Motifs 15-4

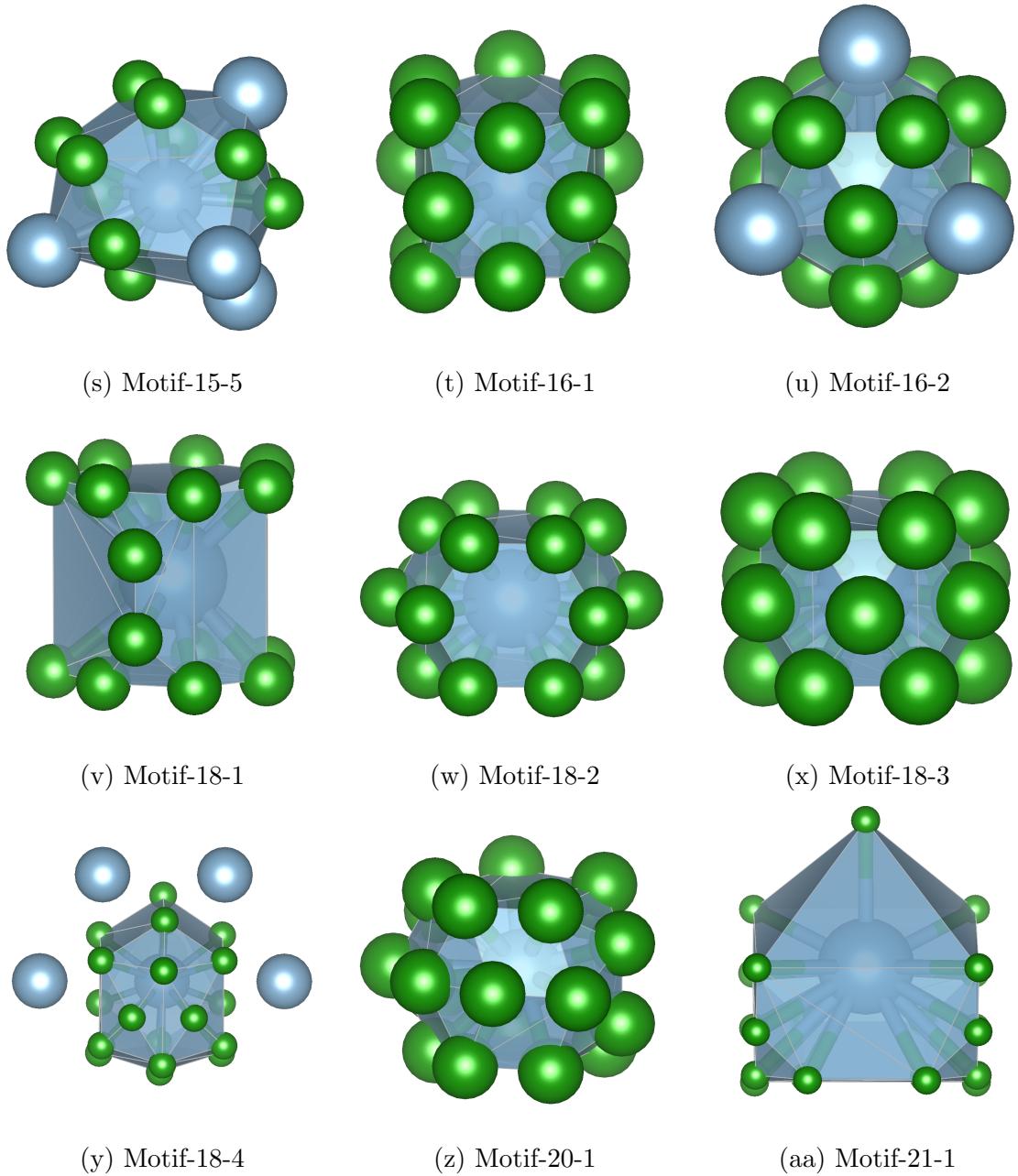


Figure S4: Motifs15-5 to Motifs21-1

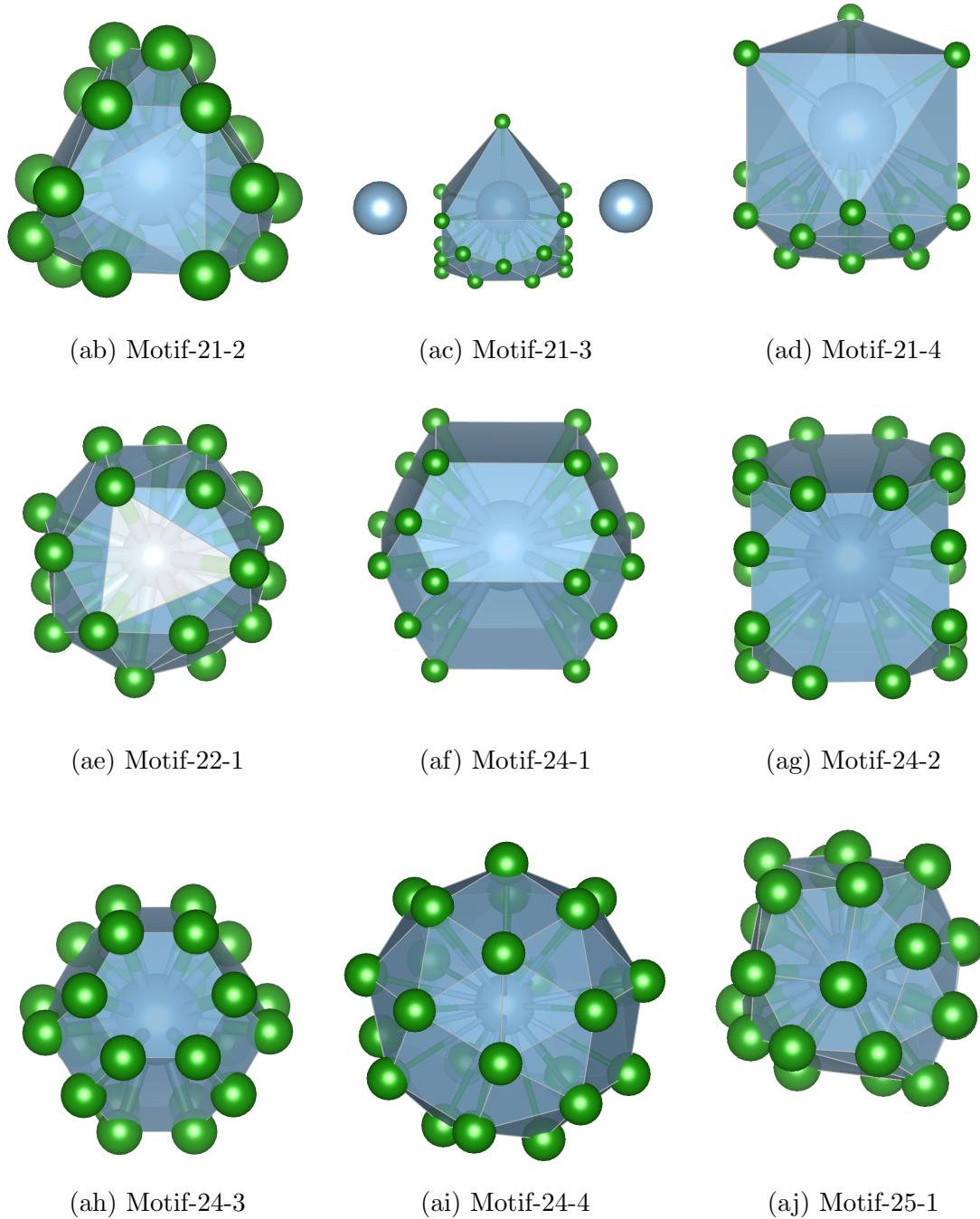


Figure S4: Motifs 21-2 to Motifs 25-1

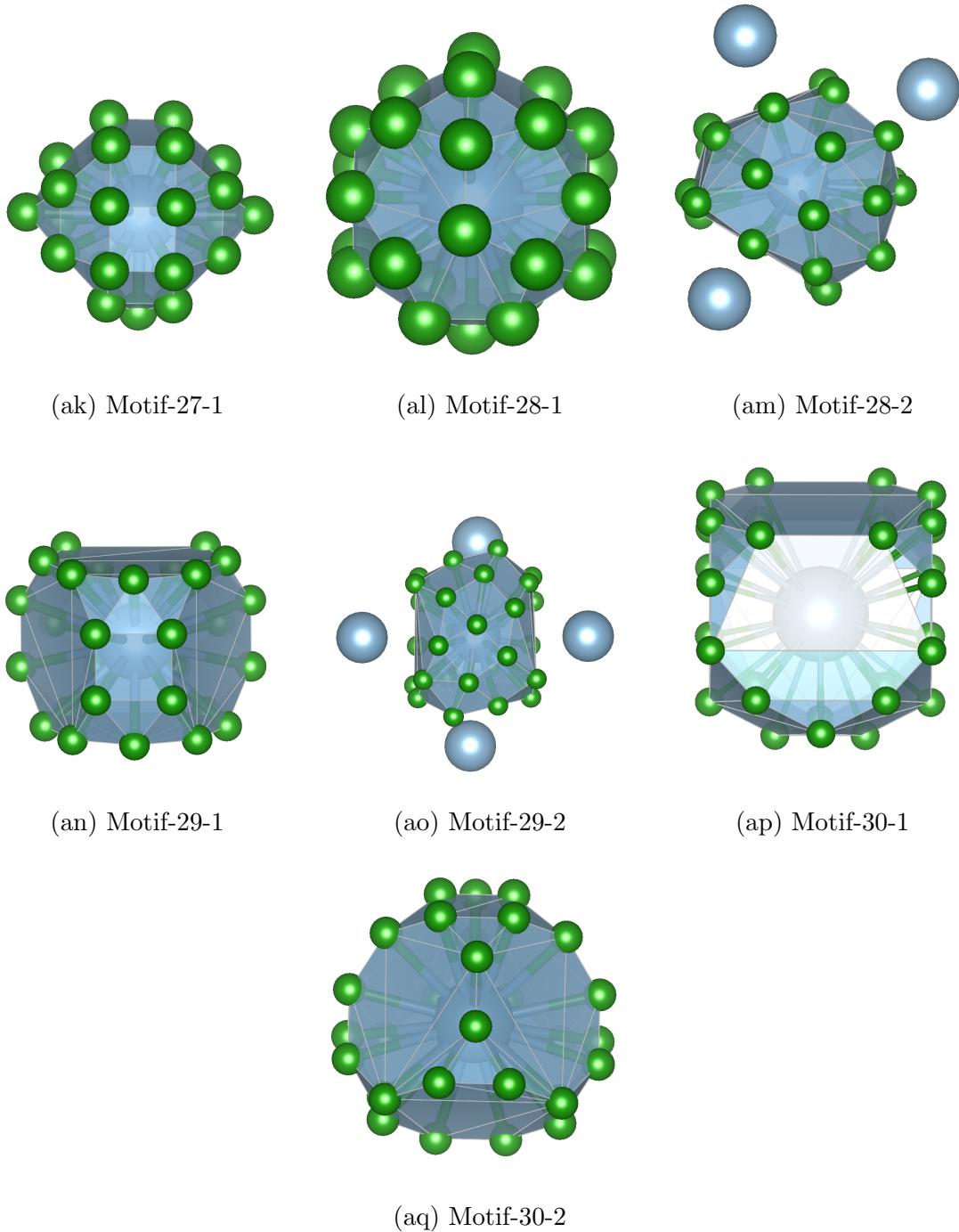
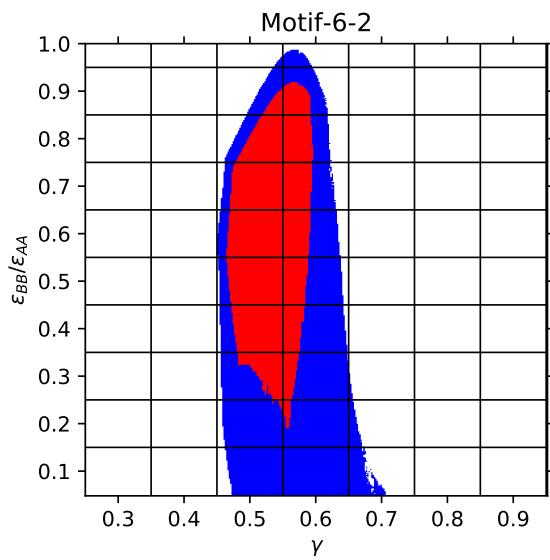
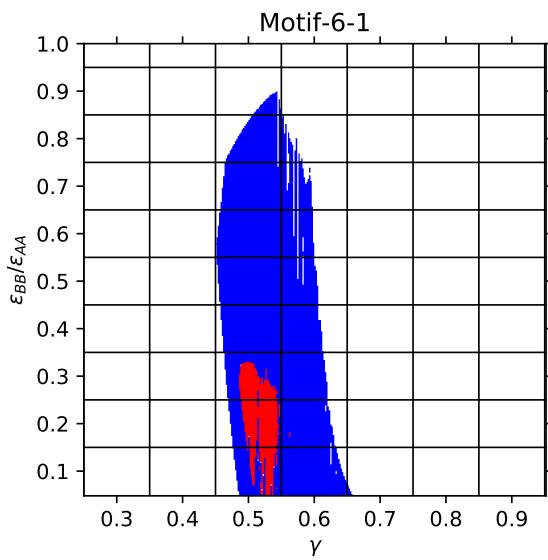
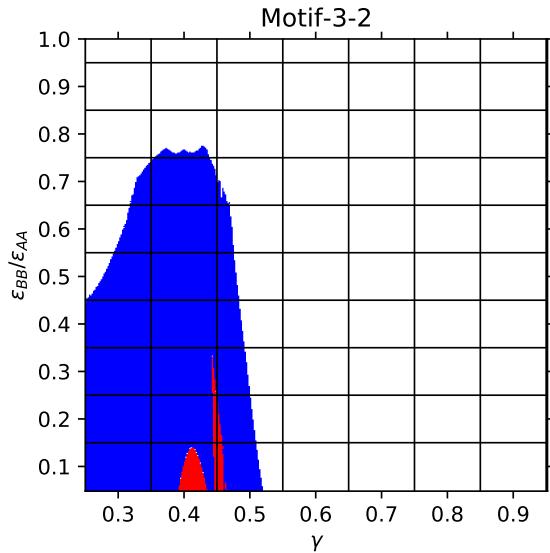
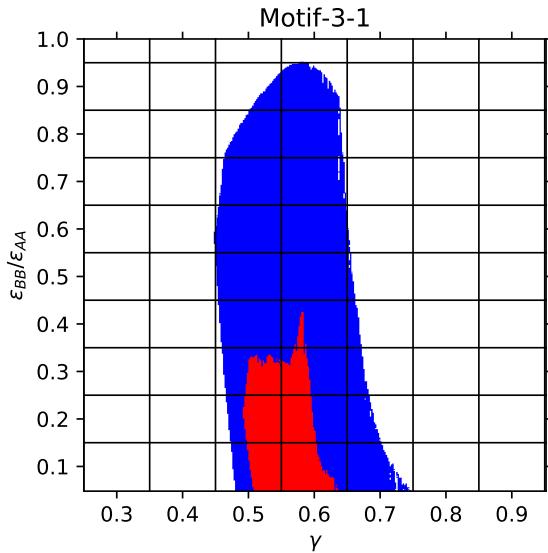
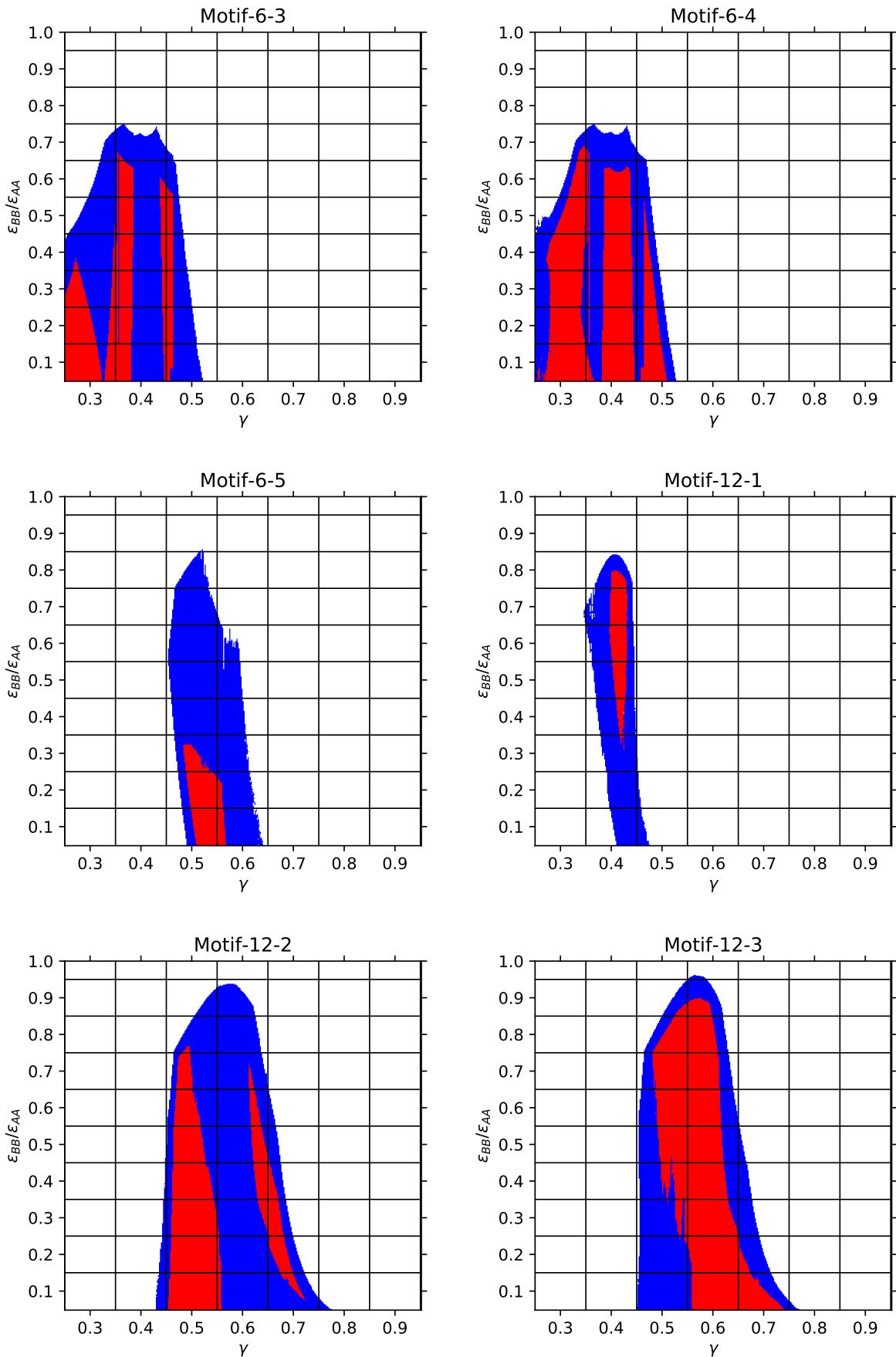
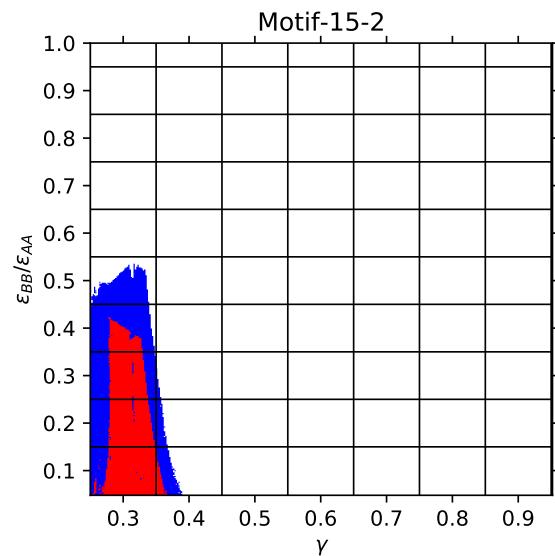
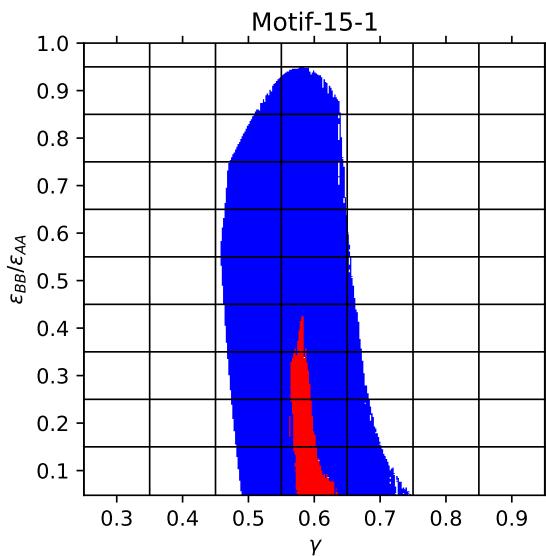
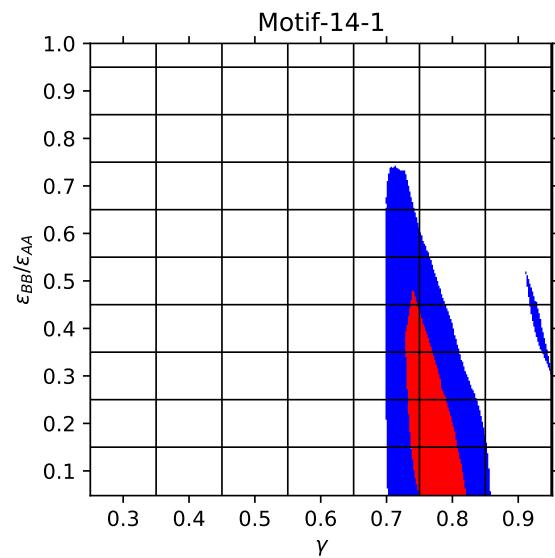
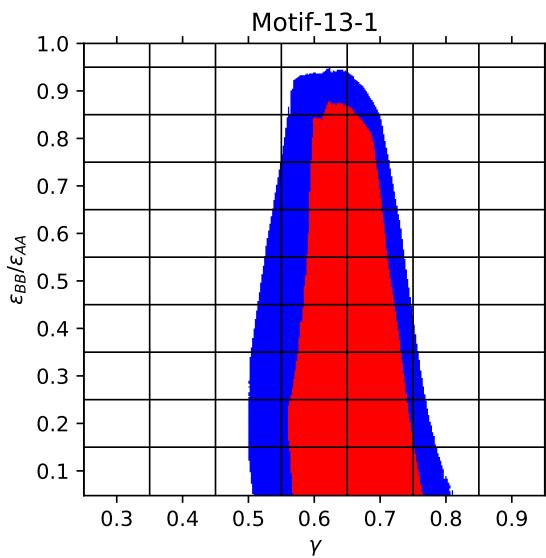
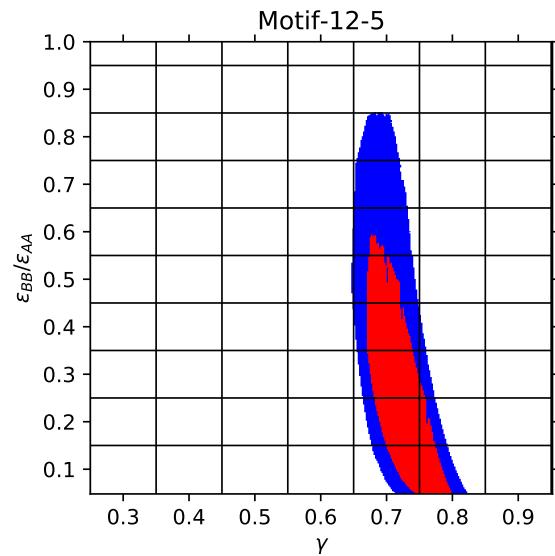
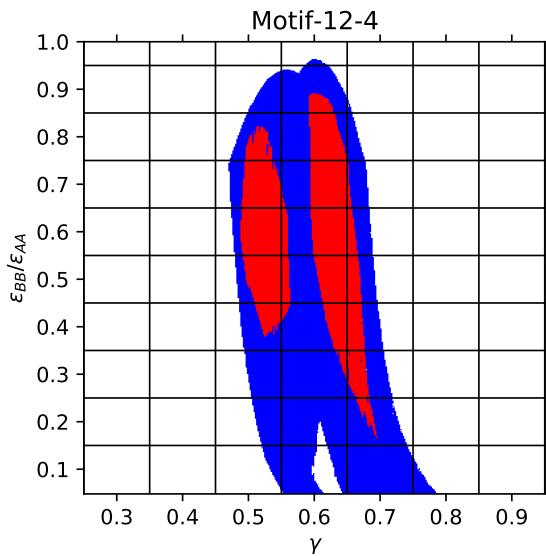


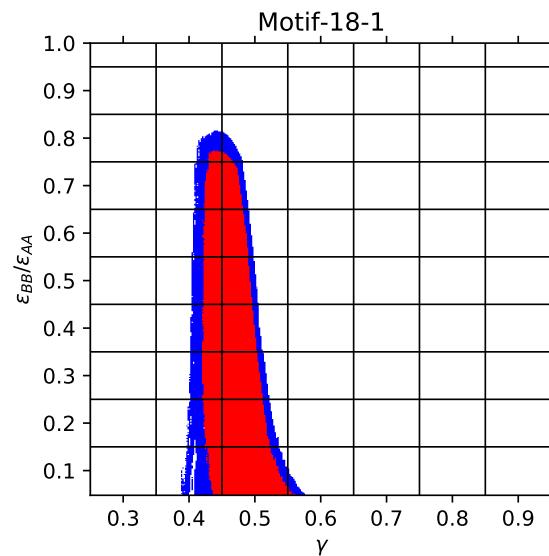
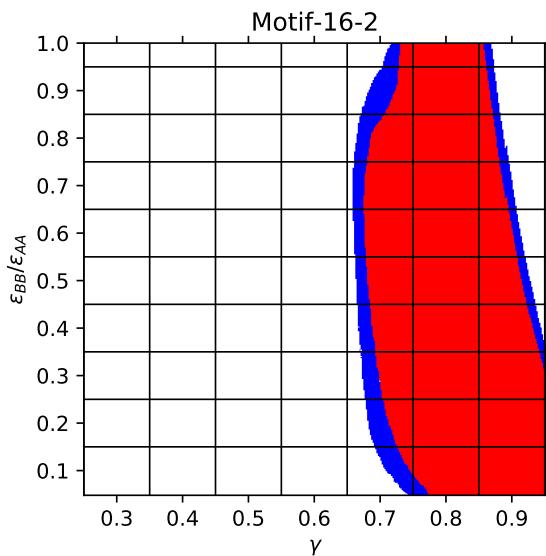
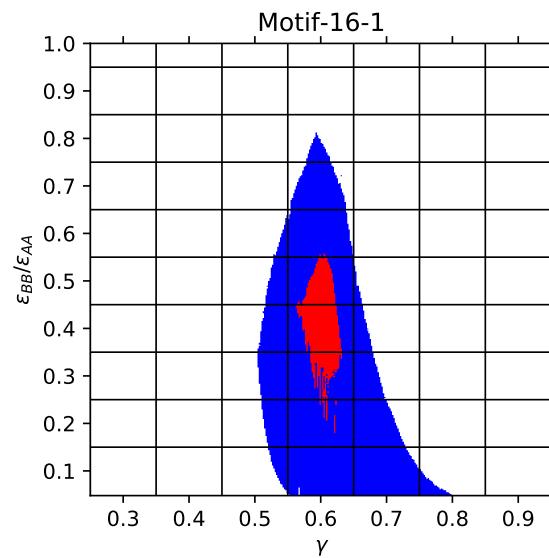
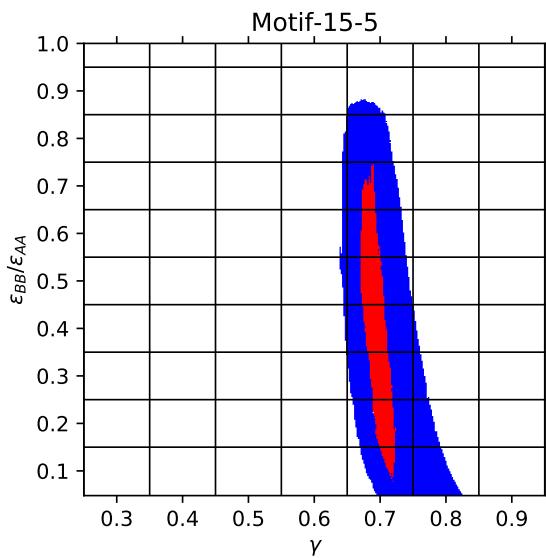
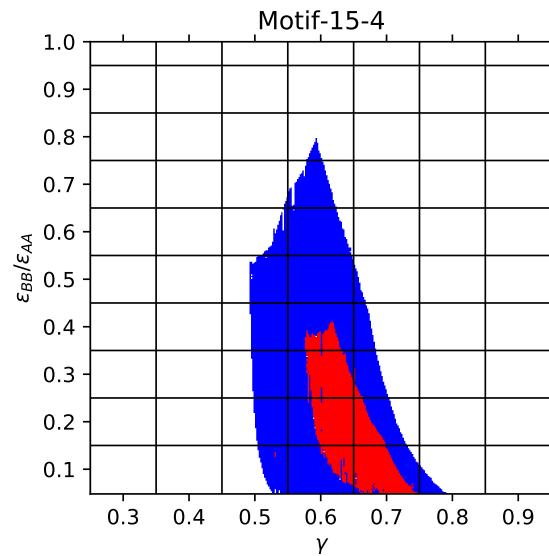
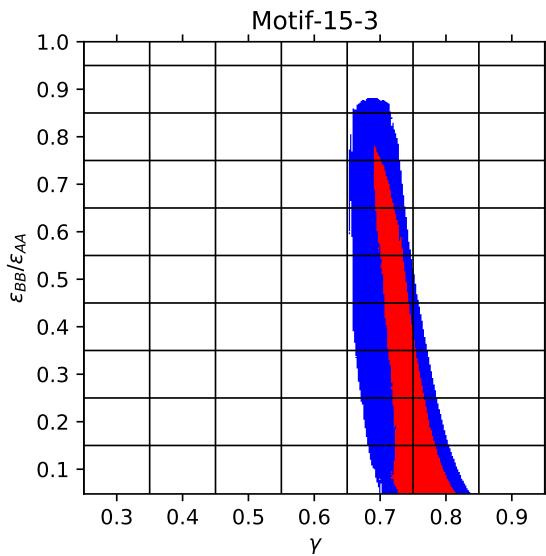
Figure S4: Motifs27-1 to Motifs30-2

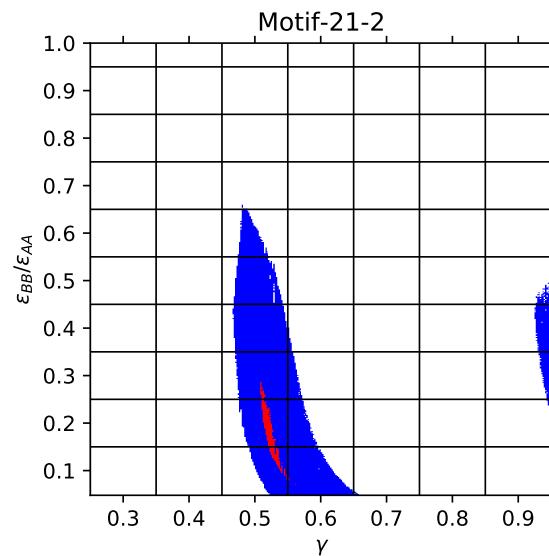
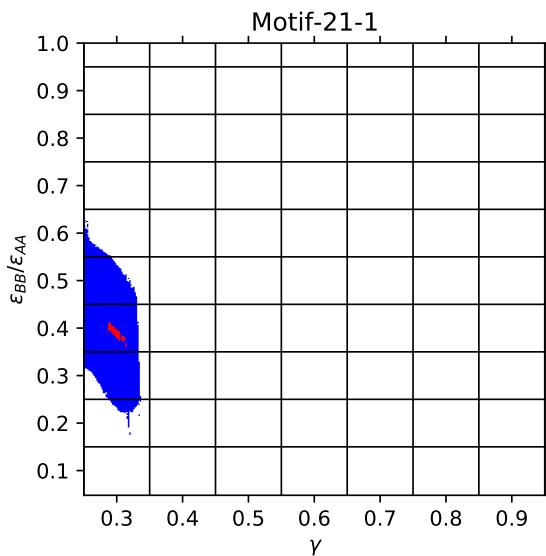
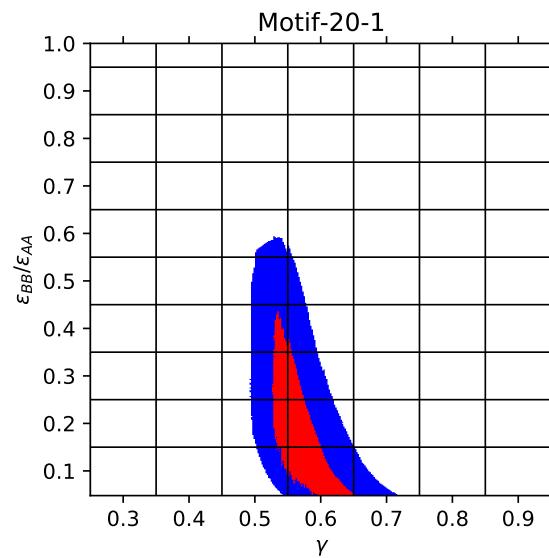
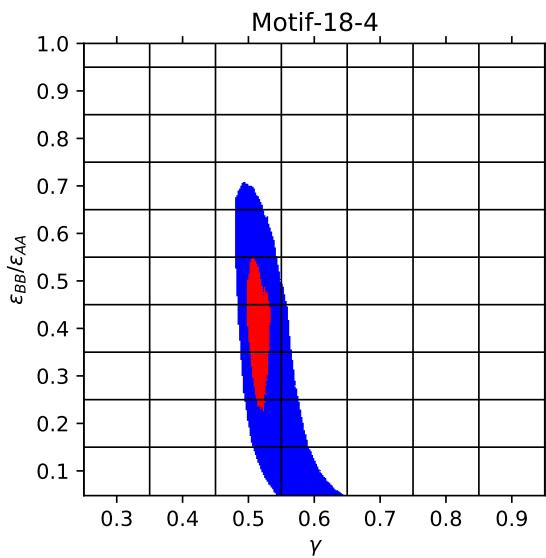
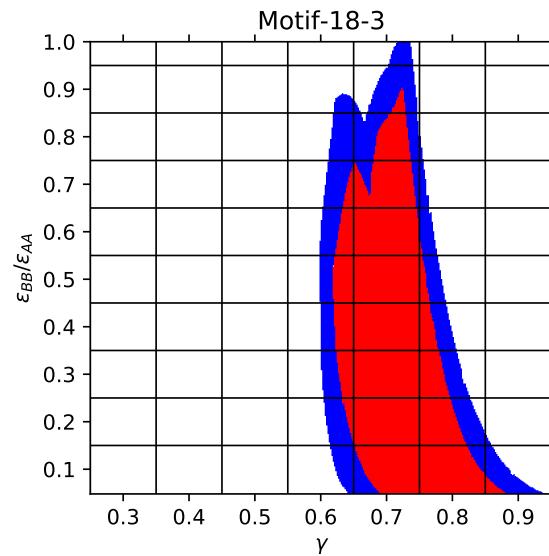
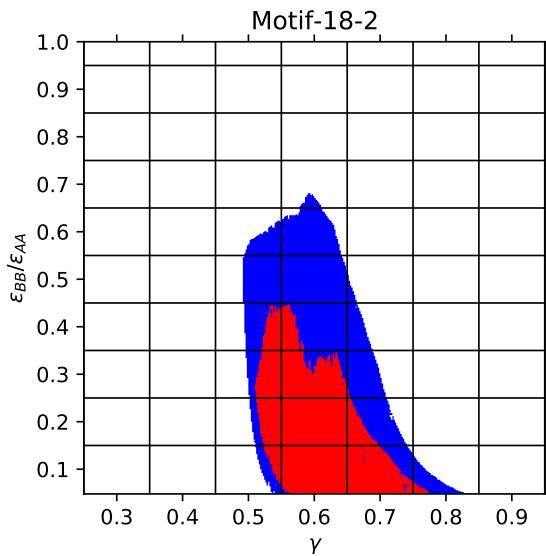
## Maps of motifs

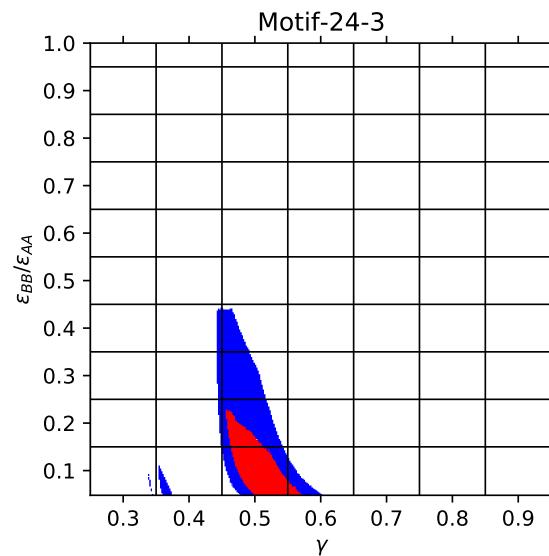
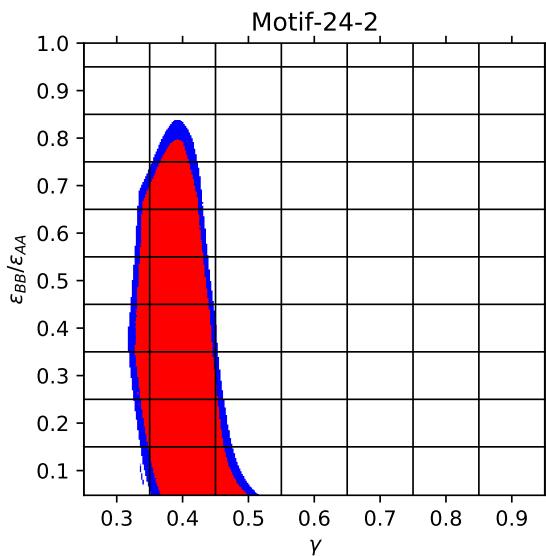
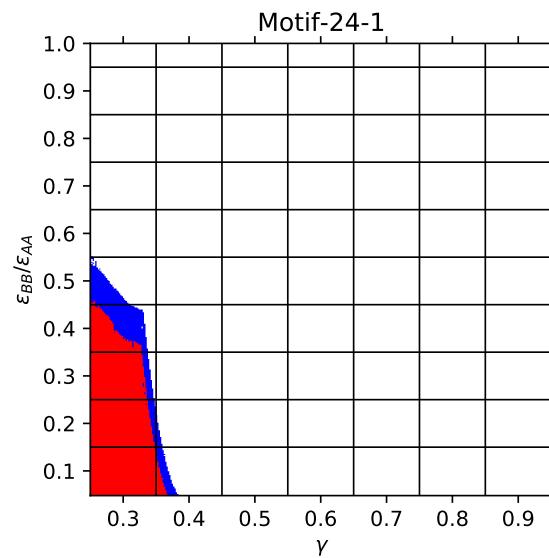
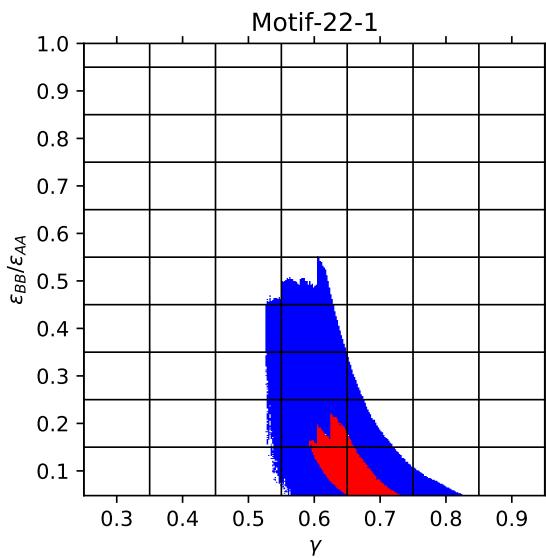
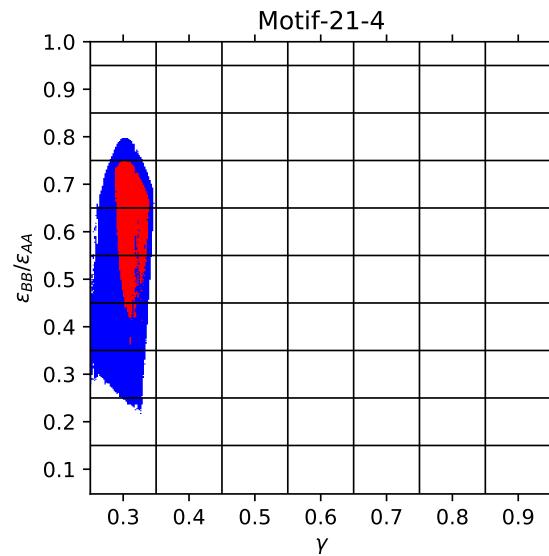
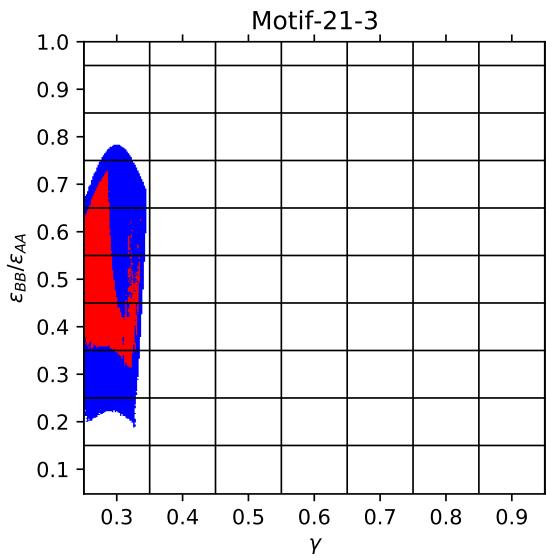


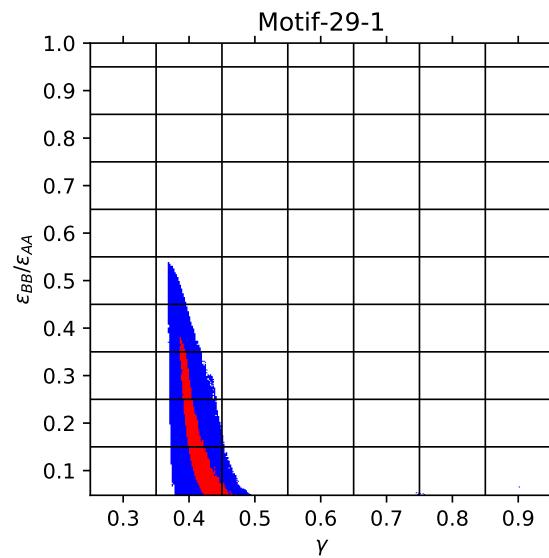
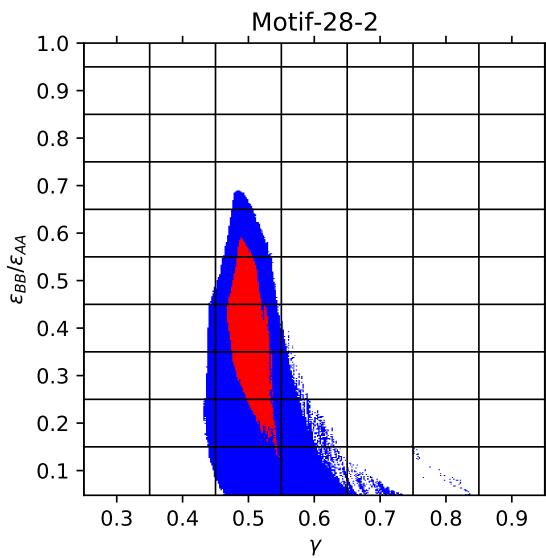
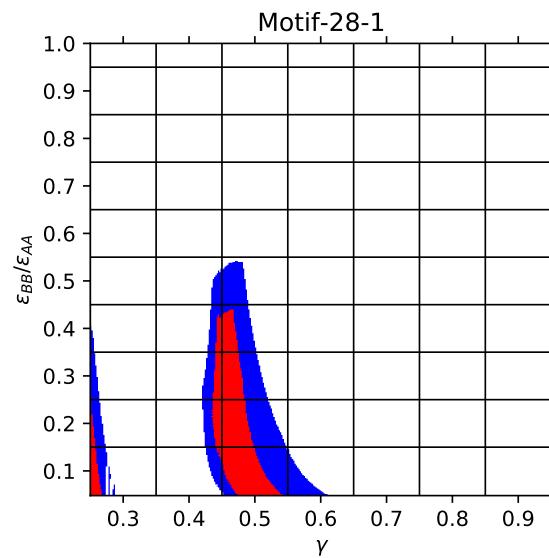
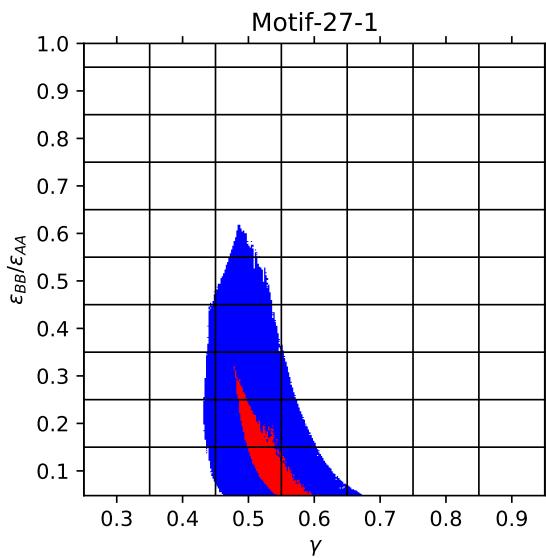
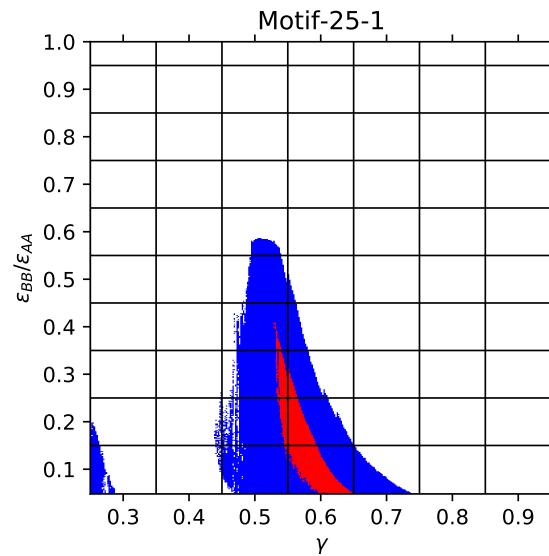
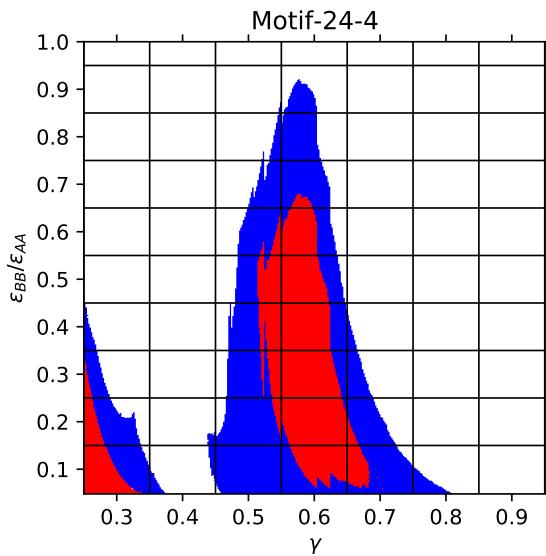


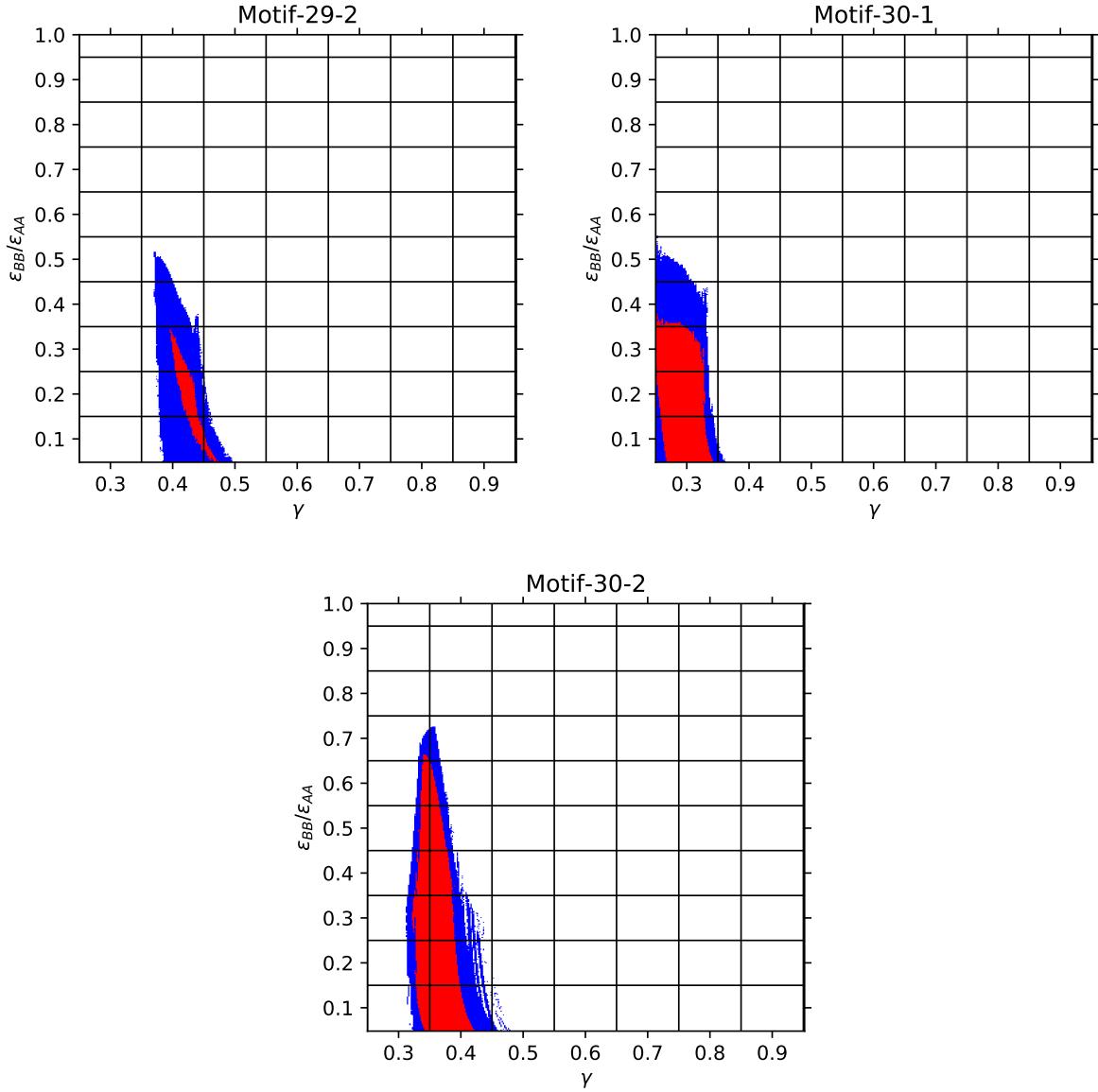












## Algorithms for motif identification and renormalized angle sequence

We illustrate the motif identification algorithm with the NaCl prototype: First we calculate the distances from the center to nearby particles and order them from small to large, see Fig. S6(a). In this way, we obtain a list of distances ( $d_i, i = 1, \dots, N, N \leq 40$ ), and we compute the ratio of distances which are associated with the current particle and the prior

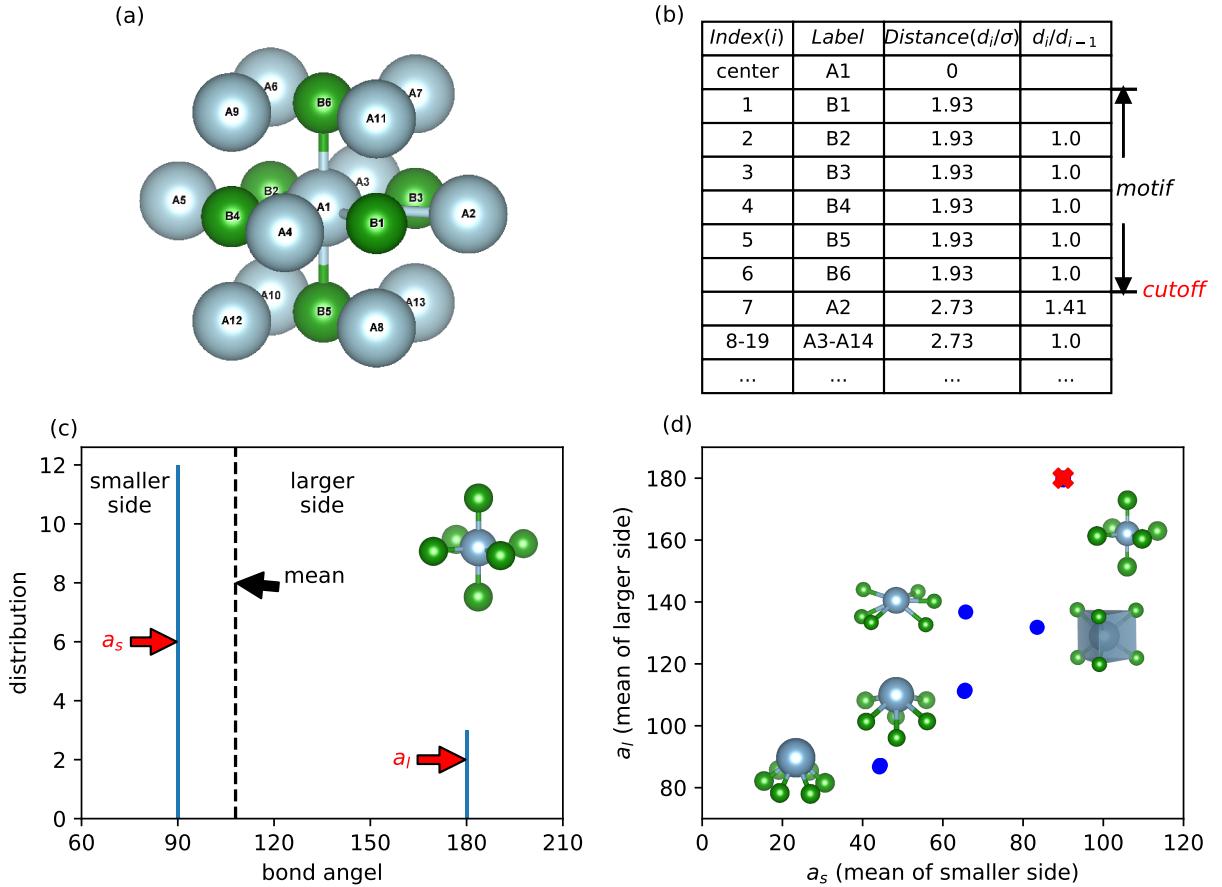


Figure S6: (a) Structure of the NaCl prototype. (b) The table of distances ( $d_i$ ) and  $d_i/d_{i-1}$ . The motif is an octahedron includes 6 B particles. (c) The distribution of bond angles and the scheme to determine RAS. (d) RASEs of 5 motifs of which the coordination number is 6.

one ( $d_i/d_{i-1}$ ), shown in Fig. S6(b). The particle  $i$  is considered as a part of the motif if the associated ratio ( $d_i/d_{i-1}$ ) is smaller than the cutoff. In our calculation, the cutoff is set to be 1.2. As shown by Fig. S6, the resulting NaCl motif is an octahedron (in green). The cutoff of 1.2 was chosen because it can differentiate the first-shells from remaining neighbors for most cases. Only in very few cases where the cluster size is large and the bond length of neighbors show variations, this cutoff may include some particles from the outer shells, which should be removed. But including these particles in the motif will not affect the conclusion that the stable and meta-stable structures share the same motif.

Having a motif, the method to identify whether it is inside a crystal or not is: Firstly,

select a center particle and take out the cluster which contains N nearest neighbors. Typically, N should be greater than the coordination number of the motif. Secondly, implement the cluster alignment method<sup>S1</sup> to compare the similarity between the motif and the cluster obtained from first step. The cutoff for the “alignment score” is set to be 0.15. A smaller alignment score indicates a higher similarity between the motif and the cluster. Thirdly, repeat the first and second step until all possibilities of center particles has been examined. In this work, all big particles (A particles) inside the unit cell are examined.

The Renormalized Angle Sequence (RAS) method, originally introduced for describing complex networks, see Ref.,<sup>S2,S3</sup> is employed to discriminate motifs with the same coordination number. It provides a simple yet powerful characterization of the motif. This descriptor is particularly relevant to identify meta-stable structures. RAS maps the motif to two featured angles. They are calculated as follows: First, we connect all particles within the motif to the center particle. Then calculate all angles between pairs of bonds, thus providing a distribution of angles. The mean of these bond angles is then calculated, see Fig. S6(c). This distribution of angles is then divided into two sets, the smaller half side and the larger half side. Then we calculate the mean of smaller side ( $a_s$ ) and the mean of larger side ( $a_l$ ). The tuple ( $a_s, a_l$ ) is the RAS descriptor. As shown in Fig. S6(d), RAS provides a very good discrimination among motifs with the same number of neighbor particles.

## Classification of Motifs by RAS

As described in Fig. S6(c-d) (and surrounding text), RAS characterizes the motif by two parameters: The average values of smaller and larger angles (i.e.  $a_s$  and  $a_l$ , respectively). In Fig. S7 (a), we plot  $a_s$  and  $a_l$  for all motifs whose CN is 6 found in stable and metastable structures, where different motifs denoted by different colors aggregate in different regions. To better show this trend, we show in Fig. S7 (b) only the regions with relatively high density of points. For instance, the block dots representing the 1.9% of other motifs are removed due to low density of points. The top five motifs are well separated in Fig. S7 (b), indicating

RAS is a reliable discriminator for CN=6 motifs. It can also be noted from Fig. S7 that for CN=6, only less than 2% of the motifs from meta-stable structures cannot be matched to any motifs from stable structures.

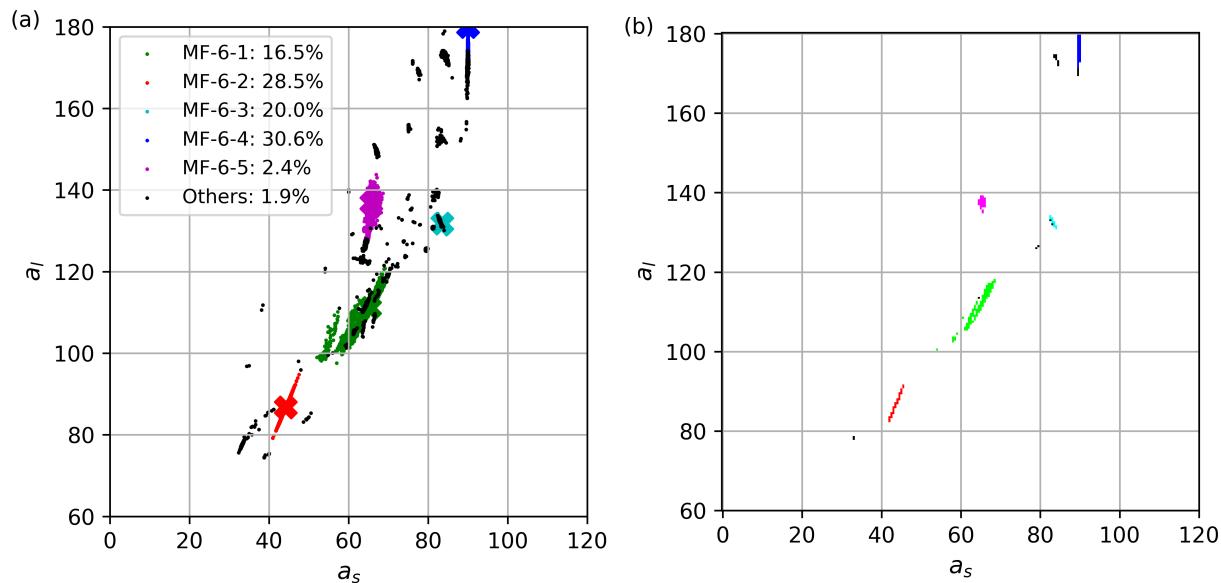


Figure S7: (a) Scatter Plot of RAS. Motifs from stable structures are marked by crosses and assigned by different colors. motifs from metastable structures are marked by points, while their color are assigned according to the similarity with the corresponding motif from stable structure. Each legend shows the percentage of motifs (from meta-stable structure) which are considered to be similar as stable structures. (b) Density Plot of RAS. Comparing with (a), the black dots disappear because of low concentration. Five different motifs are clearly separated by density plot.

## References

- [S1] Sun, Y.; Zhang, F.; Ye, Z.; Zhang, Y.; Fang, X.; Ding, Z.; Wang, C. Z.; Mendelev, M. I.; Ott, R. T.; Kramer, M. J.; Ho, K. M. Crystal Genes' in Metallic Liquids and Glasses. *Scientific Reports* **2016**, *6*, 23734.
- [S2] Lv, X.; Zhao, X.; Wu, S.; Wu, P.; Sun, Y.; Nguyen, M. C.; Shi, Y.; Lin, Z.; Wang, C.-Z.; Ho, K.-M. A Fe-P network-generation scheme to search for low-energy LiFePO<sub>4</sub> crystal structures. *J. Mater. Chem. A* **2017**, *5*, 14611–14618.

[S3] Lv, X.; Ye, Z.; Sun, Y.; Zhang, F.; Yang, L.; Lin, Z.; Wang, C. Z.; Ho, K. M. A comparative study of Sm networks in Al-10 at.%Sm glass and associated crystalline phases. *Philosophical Magazine Letters* **2018**, *98*, 27–37.