Bringing Order to Large Scale Disordered Complex Metal Alloys: $Gd_2Au_{15-x}Sb_x$ and $BaAu_xGa_{12-x}$

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Electronic Supplementary Information

Table S1. Atomic coordinates and equivalent isotropic displacement parameters ($Å^2x10^3$) for the $Gd_2Au_{10.32}Sb_{4.68}$ and $BaAu_{3.58}Ga_{8.42}$ crystals. U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

$Gd_2Au_{10.32}Sb_{4.68}$						
Gd4	*4d*	1	1/2	0	1/4	0.0095(8)
Au1	*2a*	0.403	0	0	0	0.009(1)
Sb1	*2a*	0.597	0	0	0	0.009(1)
Au2	*16m*	0.898	0.20857(9)	0.20857(9)	0.14447(6)	0.0110(3)
Sb2	*16m*	0.102	0.20857(9)	0.20857(9)	0.14447(6)	0.0110(3)
Au3	*4e*	0.405	0	0	0.3055(1)	0.0063(9)
Sb3	*4e*	0.595	0	0	0.3055(1)	0.0063(9)
Au5	*8j*	0.431	0.2941(4)	1/2	0	0.012(1)
Sb5	*8j*	0.069	0.2941(4)	1/2	0	0.012(1)
Au6	*8j*	0.184	0.1179(5)	1/2	0	0.011(1)
Sb6	*8j*	0.316	0.1179(5)	1/2	0	0.011(1)
$BaAu_{3.58}Ga_{8.42}$						
Ba1	*4a*	1	0	0	0.25	0.015(1)
Ba2	*4c*	1	0.5	0.5	0	0.019(1)
Au1	*16l*	1	0.3324(2)	0.1676(2)	0.19023(7)	0.0121(8)
Au2	*16l*	0.3(1)	0.836(3)	0.664(3)	-0.051(1)	0.010(6)
Ga2	*16l*	0.2(2)	0.836(3)	0.664(3)	-0.051(1)	0.010(6)
Au3	*16l*	0.05(2)	0.871(9)	0.629(9)	-0.044(1)	0.03(1)
Ga3	*16l*	0.45(2)	0.871(9)	0.629(9)	-0.044(1)	0.03(1)
Au4	*16l*	0.25(3)	0.6678(6)	0.8322(6)	-0.0693(3)	0.019(3)
Ga4	*16l*	0.25(3)	0.6678(6)	0.8322(6)	-0.0693(3)	0.019(3)
Au5	*16l*	0.01(3)	0.634(1)	0.866(1)	-0.0342(9)	0.037(9)
Ga5	*16l*	0.49(3)	0.634(1)	0.866(1)	-0.0342(9)	0.037(9)
Au6	*32m*	0.09(2)	0.0535(4)	0.2006(4)	0.1286(1)	0.016(2)
Ga6	*32m*	0.91(2)	0.0535(4)	0.2006(4)	0.1286(1)	0.016(2)
Ga7	*16l*	1	0.3789(4)	0.1211(4)	0.2889(2)	0.0121(8)

Table S2. Interatomic distance data for all samples. Exceptionally short distances are products of split positions and discussed further in the text.

$Gd_2Au_{11.43}Sb_{3.57}$

Au1—Sb2^{i}	2.973 (3)	Au5—Au6	1.289 (5)
Au1—Au2^{i}	2.973 (3)	Au5—Sb5^{xii}	2.131 (5)
Au1—Sb2^{ii}	2.973 (3)	Au5—Au5^{xii}	2.131 (5)
Au1—Sb2^{iii}	2.973 (3)	Au5—Sb5^{viii}	2.131 (5)
Au1—Au2^{iv}	2.973 (3)	Au5—Au5^{viii}	2.131 (5)
Au1—Au2^{v}	2.973 (3)	Au5—Sb5^{xiii}	3.013 (8)
Au1—Sb2^{v}	2.973 (3)	Au5—Au5^{xiii}	3.013 (8)
Au1—Sb2^{iv}	2.973 (3)	Au5—Sb2^{vii}	3.015 (3)
Au1—Au2^{iii}	2.973 (3)	Au5—Au2^{vii}	3.015 (3)
Au1—Au2^{ii}	2.973 (3)	Au6—Sb6^{xiv}	1.738 (9)
Au1—Au2^{vi}	2.973 (3)	Au6—Au6^{xiv}	1.738 (9)
Au1—Sb2^{vii}	2.973 (3)	Au6—Sb2^{xv}	3.023 (3)
Au2—Sb5^{viii}	3.015 (3)	Au6—Au2^{xv}	3.023 (3)
Au2—Au5^{viii}	3.015 (3)	Au6—Au2^{vii}	3.023 (3)
Au2—Au5	3.015 (3)	Au6—Sb2^{vii}	3.023 (3)
Au2—Gd4	3.020 (3)	Au6—Au2^{xii}	3.023 (3)
Au2—Gd4^{ix}	3.020 (3)	Au6—Sb2^{xii}	3.023 (3)
Au2—Au6	3.023 (3)	Au3—Sb2^{ix}	3.098 (4)
Au2—Sb6^{viii}	3.023 (3)	Au3—Au2^{ix}	3.098 (4)
Au2—Au6^{viii}	3.023 (3)	Au3—Sb2^{xvi}	3.098 (4)
Gd4—Sb2^{iii}	3.020 (3)	Au3—Au2^{xvi}	3.098 (4)
Gd4—Sb2^{x}	3.020 (3)	Au3—Sb2^{xi}	3.098 (4)
Gd4—Au2^{iii}	3.020 (3)	Au3—Au2^{xvii}	3.098 (4)
Gd4—Au2^{x}	3.020 (3)	Au3—Sb2^{xvii}	3.098 (4)
Gd4—Sb2^{xi}	3.020 (3)	Au3—Au2^{xi}	3.098 (4)

Gd4—Au2^{viii}	3.020 (3)	Au3—Sb5^{xvii}	3.128 (4)
Gd4—Au2^{ix}	3.020 (3)	Au3—Sb5^{ix}	3.128 (4)
Gd4—Au2^{xi}	3.020 (3)	Au3—Au5^{xvii}	3.128 (4)
Gd4—Sb2^{ix}	3.020 (3)	Au3—Au5^{ix}	3.128 (4)
Gd4—Sb2^{viii}	3.020 (3)		

$Gd_2Au_{10.32}Sb_{4.68}$

Au1—Au2	2.9607 (14)	Gd4—Au2^{xix}	3.009 (2)
Au1—Sb2^{i}	2.9607 (14)	Au5—Au6	1.287 (5)
Au1—Au2^{i}	2.9607 (14)	Au5—Sb5^{xii}	2.128 (4)
Au1—Sb2^{ii}	2.9607 (14)	Au5—Au5^{xii}	2.128 (4)
Au1—Au2^{v}	2.9607 (14)	Au5—Sb5^{viii}	2.128 (4)
Au1—Sb2^{iii}	2.9607 (14)	Au5—Au5^{viii}	2.128 (4)
Au1—Sb2^{v}	2.9607 (14)	Au5—Au2^{vii}	3.008 (2)
Au1—Au2^{iii}	2.9607 (14)	Au5—Sb2^{vii}	3.008 (2)
Au1—Au2^{ii}	2.9607 (14)	Au5—Au2^{xv}	3.008 (2)
Au1—Au2^{iv}	2.9607 (14)	Au5—Sb2^{xv}	3.008 (2)
Au1—Sb2^{iv}	2.9607 (14)	Au3—Sb2^{ix}	3.092 (2)
Au2—Sb5^{viii}	3.0075 (10)	Au3—Au2^{ix}	3.092 (2)
Au2—Au5^{viii}	3.0075 (10)	Au3—Sb2^{xvi}	3.092 (2)
Au2—Au5	3.008 (2)	Au3—Au2^{xvi}	3.092 (2)
Au2—Gd4	3.0092 (12)	Au3—Sb2^{xi}	3.092 (2)
Au2—Gd4^{ix}	3.009 (2)	Au3—Au2^{xvii}	3.092 (2)
Au2—Sb6^{viii}	3.0155 (11)	Au3—Sb2^{xvii}	3.092 (2)
Au2—Au6^{viii}	3.0155 (11)	Au3—Au2^{xi}	3.092 (2)
Au2—Au6	3.016 (2)	Au3—Sb5^{xvii}	3.119 (2)
Gd4—Sb2^{iii}	3.0092 (12)	Au3—Sb5^{ix}	3.119 (2)
Gd4—Sb2^{x}	3.009 (2)	Au3—Au5^{xvii}	3.119 (2)
Gd4—Au2^{iii}	3.0092 (12)	Au3—Au5^{ix}	3.119 (2)

Gd4—Au2^{x}	3.009 (2)	Au6—Sb6^{xiv}	1.723 (7)
Gd4—Sb2^{xi}	3.009 (2)	Au6—Au6^{xiv}	1.723 (7)
Gd4—Au2^{viii}	3.0092 (12)	Au6—Sb5^{xiv}	3.011 (4)
Gd4—Sb2^{viii}	3.0092 (12)	Au6—Au5^{xiv}	3.011 (4)
Gd4—Sb2^{ix}	3.009 (2)	Au6—Au2^{xv}	3.016 (2)
Gd4—Au2^{ix}	3.009 (2)	Au6—Sb2^{xv}	3.016 (2)
Gd4—Au2^{xi}	3.009 (2)	Au6—Sb2^{vii}	3.016 (2)
Gd4—Au2^{xviii}	3.0092 (12)	Au6—Au2^{vii}	3.016 (2)

$BaAu_{4.53}Ga_{7.47}$

Au1—Ga8^{x}	2.629 (4)	Ga4—Au4^{vii}	2.553 (17)
Au1—Ga8^{xi}	2.629 (4)	Ga4—Au5	2.65 (2)
Au1—Ga8	2.632 (6)	Ga4—Au5^{xv}	2.658 (14)
Au1—Ga8^{ix}	2.662 (6)	Ga4—Au5^{xxv}	2.658 (14)
Au1—Ga6^{iii}	2.783 (3)	Ga4—Ga6^{xxiv}	2.724 (17)
Au1—Au6^{iii}	2.783 (3)	Ga4—Au6^{xxiv}	2.724 (17)
Au1—Ga6^{xx}	2.783 (3)	Ga4—Ga6^{xiii}	2.724 (17)
Au1—Au6^{xx}	2.783 (3)	Ga4—Au6^{xiii}	2.724 (17)
Au1—Au6	2.959 (4)	Au5—Ga5	1.064 (14)
Au1—Au6^{xxi}	2.959 (4)	Au5—Ga4^{xxvi}	2.658 (14)
Au1—Ga6^{xxi}	2.959 (4)	Au5—Ga4^{viii}	2.658 (14)
Ba2—Au1^{vi}	3.6152 (10)	Au5—Ga5^{vii}	2.674 (15)
Ba2—Au1^{ix}	3.6152 (10)	Au5—Ga6^{xxvii}	2.733 (5)
Ba2—Au1^{xi}	3.6152 (10)	Au5—Au6^{xxvii}	2.733 (5)
Ba2—Au1^{iii}	3.6152 (10)	Au5—Ga6^{xxviii}	2.733 (5)
Ba2—Au1^{xvi}	3.6152 (10)	Au5—Au6^{xxviii}	2.733 (5)
Ba2—Au1^{xvii}	3.6152 (10)	Au5—Ga6^{xxiv}	2.919 (6)
Ba2—Au1^{iv}	3.6152 (10)	Au5—Au6^{xxiv}	2.919 (6)
Ba2—Ga8	3.631 (3)	Ga5—Ga5^{vii}	1.68 (3)

Ba2—Ga8^{xvi}	3.631 (3)	Ga5—Ga4^{viii}	2.253 (17)
Ba2—Ga8^{iv}	3.631 (3)	Ga5—Ga4^{xxvi}	2.253 (17)
Ba2—Ga8^{ix}	3.631 (3)	Ga5—Au4^{viii}	2.635 (11)
Ba3—Au4	3.535 (3)	Ga5—Au4^{xxvi}	2.635 (11)
Ba3—Au4^{vii}	3.535 (3)	Ga5—Au5^{vii}	2.674 (15)
Ba3—Au4^{xxii}	3.535 (3)	Ga5—Ga4^{xxix}	3.01 (2)
Ba3—Au4^{xxiii}	3.535 (3)	Ga5—Ga4^{xxiii}	3.01 (2)
Ba3—Au4^{xv}	3.535 (3)	Ga5—Ba3^{xxx}	3.547 (6)
Ba3—Au4^{xiii}	3.535 (3)	Au6—Au4^{xiii}	2.543 (6)
Ba3—Au4^{xii}	3.535 (3)	Au6—Ga6^{iv}	2.556 (5)
Ba3—Au4^{viii}	3.535 (3)	Au6—Au6^{iv}	2.556 (5)
Ba3—Ga5^{vii}	3.547 (6)	Au6—Ga6^{iii}	2.556 (5)
Ba3—Ga5^{xxii}	3.547 (6)	Au6—Au6^{iii}	2.556 (5)
Ba3—Ga5^{xiii}	3.547 (6)	Au6—Ga4^{xiii}	2.723 (17)
Ba3—Ga5^{xii}	3.547 (6)	Au6—Au5^{xxxi}	2.733 (5)
Au4—Ga4	0.509 (16)	Au6—Ga8^{ix}	2.739 (6)
Au4—Au5	2.144 (10)	Au6—Au1^{iv}	2.783 (3)
Au4—Ga6^{xxiv}	2.543 (6)	Au6—Au5^{xiii}	2.919 (6)
Au4—Au6^{xxiv}	2.543 (6)	Au6—Au6^{xxi}	3.083 (7)
Au4—Ga6^{xiii}	2.543 (6)	Ga8—Au1^{x}	2.629 (4)
Au4—Au6^{xiii}	2.543 (6)	Ga8—Au1^{xi}	2.629 (4)
Au4—Ga4^{vii}	2.553 (17)	Ga8—Au1^{ix}	2.662 (6)
Au4—Ga5	2.610 (16)	Ga8—Ga6^{ix}	2.739 (6)
Au4—Ga5^{xxv}	2.635 (11)	Ga8—Au6^{ix}	2.739 (6)
Au4—Ga5^{xv}	2.635 (11)	Ga8—Ga6^{xxxii}	2.739 (6)
Au4—Au4^{vii}	2.640 (13)	Ga8—Au6^{xxxii}	2.739 (6)
Au4—Au5^{xv}	2.946 (7)	Ga8—Ga8^{xi}	2.955 (10)
Ga4—Ga5^{xxv}	2.253 (17)	Ga8—Ga8^{x}	2.955 (10)
Ga4—Ga5^{xv}	2.253 (17)	Ga8—Ba2^{ix}	3.631 (3)

Ga4—Ga4^{vii}	2.37 (4)		

$BaAu_{3.58}Ga_{8.42}$

Ba1—Au1^{i}	3.619 (3)	Au3—Ga5^{xvii}	2.32 (8)
Ba1—Au1^{ii}	3.619 (3)	Au3—Au5^{xvii}	2.32 (8)
Ba1—Au1^{iii}	3.619 (3)	Au3—Ga5^{xviii}	2.32 (8)
Ba1—Au1^{iv}	3.619 (3)	Au3—Au5^{xviii}	2.32 (8)
Ba1—Au1^{v}	3.619 (3)	Au3—Ga2^{viii}	2.53 (4)
Ba1—Au1^{vi}	3.619 (3)	Au3—Au2^{viii}	2.53 (4)
Ba1—Au1^{vii}	3.619 (3)	Au3—Au4	2.60 (12)
Ba1—Au1	3.620 (3)	Au3—Ga4^{xviii}	2.71 (7)
Ba1—Ga7^{v}	3.634 (4)	Au3—Au4^{xviii}	2.71 (7)
Ba1—Ga7	3.634 (4)	Au4—Au5	1.008 (19)
Ba1—Ga7^{vi}	3.634 (4)	Au4—Ga3^{xix}	2.71 (7)
Ba1—Ga7^{i}	3.634 (4)	Au4—Au3^{xix}	2.71 (7)
Ba2—Au5	3.534 (8)	Au4—Ga3^{xii}	2.71 (7)
Ba2—Ga5^{viii}	3.534 (8)	Au4—Au3^{xii}	2.71 (7)
Ba2—Ga5^{ix}	3.534 (8)	Au4—Ga5^{viii}	2.74 (3)
Ba2—Au5^{viii}	3.534 (8)	Au4—Au5^{viii}	2.74 (3)
Ba2—Au5^{ix}	3.534 (8)	Au5—Ga5^{viii}	1.79 (5)
Ba2—Ga5^{x}	3.534 (8)	Au5—Au5^{viii}	1.79 (5)
Ba2—Au5^{x}	3.534 (8)	Au5—Ga3^{xii}	2.32 (8)
Ba2—Ga5^{xi}	3.534 (8)	Au5—Au3^{xii}	2.32 (8)
Ba2—Ga5^{xii}	3.534 (8)	Au5—Ga3^{xix}	2.32 (8)
Ba2—Au5^{xi}	3.534 (8)	Au5—Au3^{xix}	2.32 (8)
Ba2—Au5^{xii}	3.534 (8)	Au5—Au2^{xii}	2.66 (3)
Au1—Ga7^{xiii}	2.621 (4)	Au5—Ga2^{xii}	2.66 (3)
Au1—Ga7^{iii}	2.621 (4)	Au6—Ga2^{x}	2.54 (3)
Au1—Ga7	2.643 (6)	Au6—Au2^{x}	2.54 (3)

Au1—Ga7^{ii}	2.678 (6)	Au6—Ga6^{vii}	2.575 (5)
Au1—Ga6^{iv}	2.773 (4)	Au6—Au6^{vii}	2.575 (5)
Au1—Au6^{iv}	2.773 (4)	Au6—Ga6^{iv}	2.575 (5)
Au1—Ga6^{xiv}	2.773 (4)	Au6—Au6^{iv}	2.575 (5)
Au1—Au6^{xiv}	2.773 (4)	Au6—Ga7^{ii}	2.727 (6)
Au1—Ga6^{xv}	2.943 (4)	Au6—Ga4^{xx}	2.740 (6)
Au1—Au6^{xv}	2.943 (4)	Au6—Au4^{xx}	2.740 (6)
Au1—Au6	2.943 (4)	Au6—Au3^{x}	2.74 (4)
Au2—Au4	2.14 (4)	Au6—Ga3^{x}	2.74 (4)
Au2—Ga3^{viii}	2.53 (4)	Au6—Au1^{vii}	2.773 (4)
Au2—Au3^{viii}	2.53 (4)	Ga7—Au1^{xiii}	2.621 (4)
Au2—Ga6^{xvi}	2.54 (3)	Ga7—Au1^{iii}	2.621 (4)
Au2—Au6^{xvi}	2.54 (3)	Ga7—Au1^{ii}	2.678 (6)
Au2—Ga6^{x}	2.54 (3)	Ga7—Ga6^{ii}	2.727 (6)
Au2—Au6^{x}	2.54 (3)	Ga7—Au6^{ii}	2.727 (6)
Au2—Au5	2.54 (3)	Ga7—Ga6^{xxi}	2.727 (6)
Au2—Ga5^{xvii}	2.66 (3)	Ga7—Au6^{xxi}	2.727 (6)
Au2—Au5^{xvii}	2.66 (3)	Ga7—Ga7^{iii}	2.941 (10)
Au3—Ga3^{viii}	2.32 (7)	Ga7—Ga7^{xiii}	2.941 (10)
Au3—Au3^{viii}	2.32 (7)	Ga7—Ga7^{xxii}	3.003 (11)

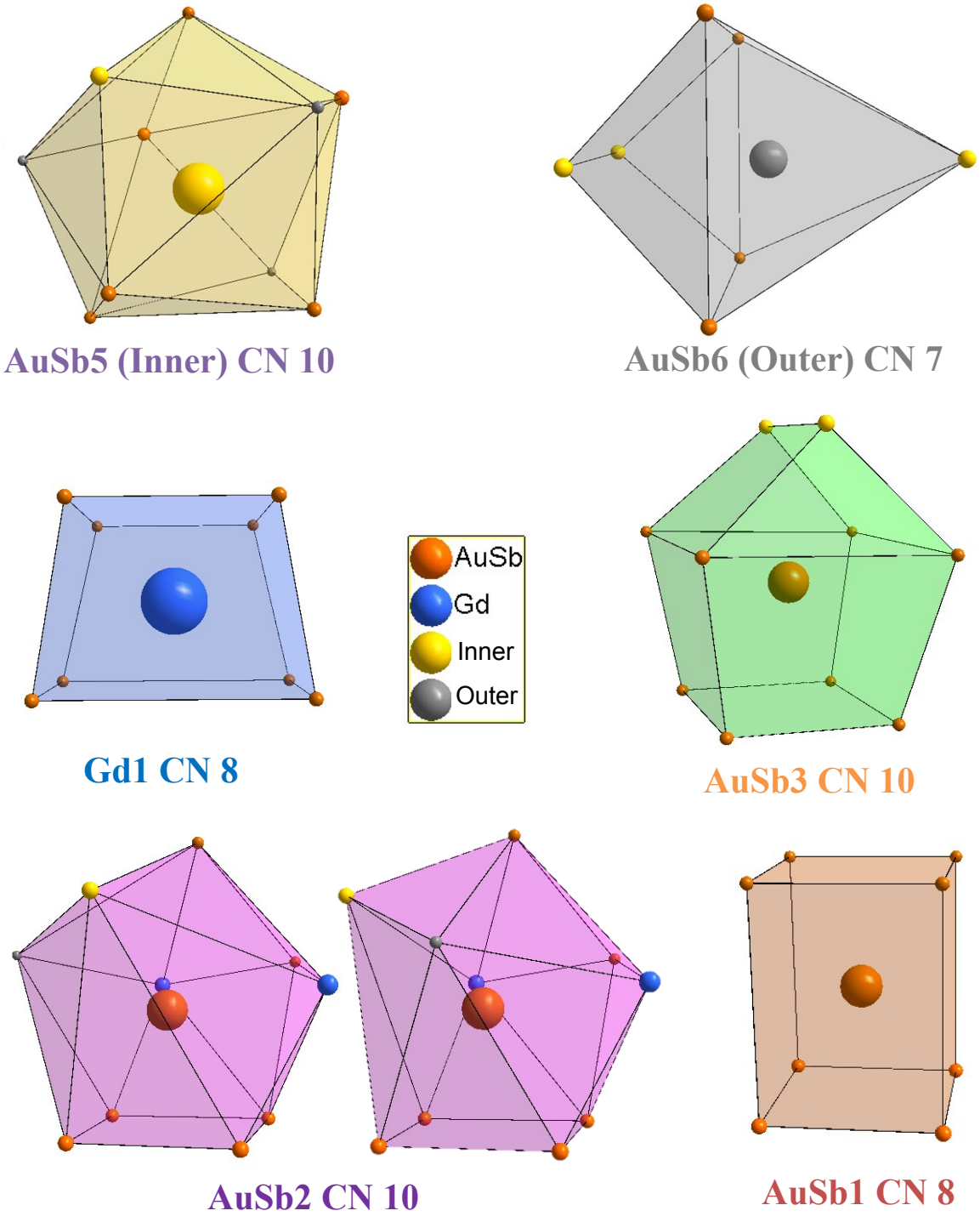


Figure S1. Coordination polyhedra for each site are illustrated above. For clarity, disordered positions that would be too close to the central atom to also be occupied have been removed from the top two polyhedra. The (AuSb)2 polyhedra form enantiomorphs depending on the inner and outer site fillings.

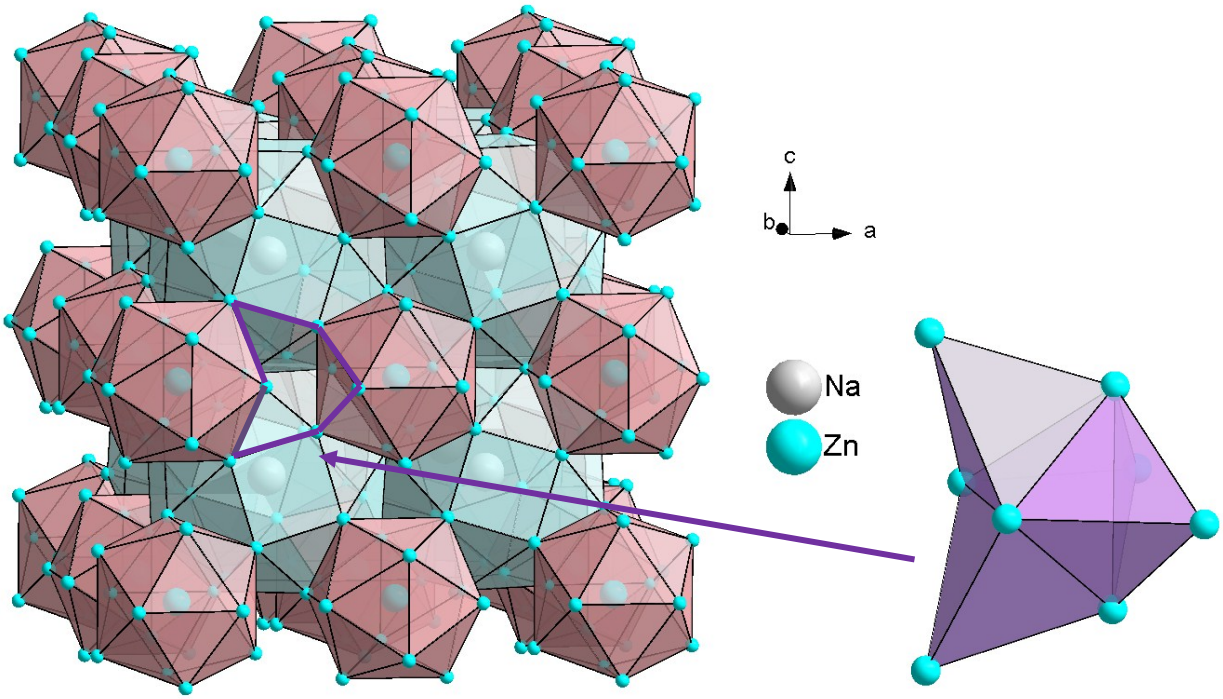
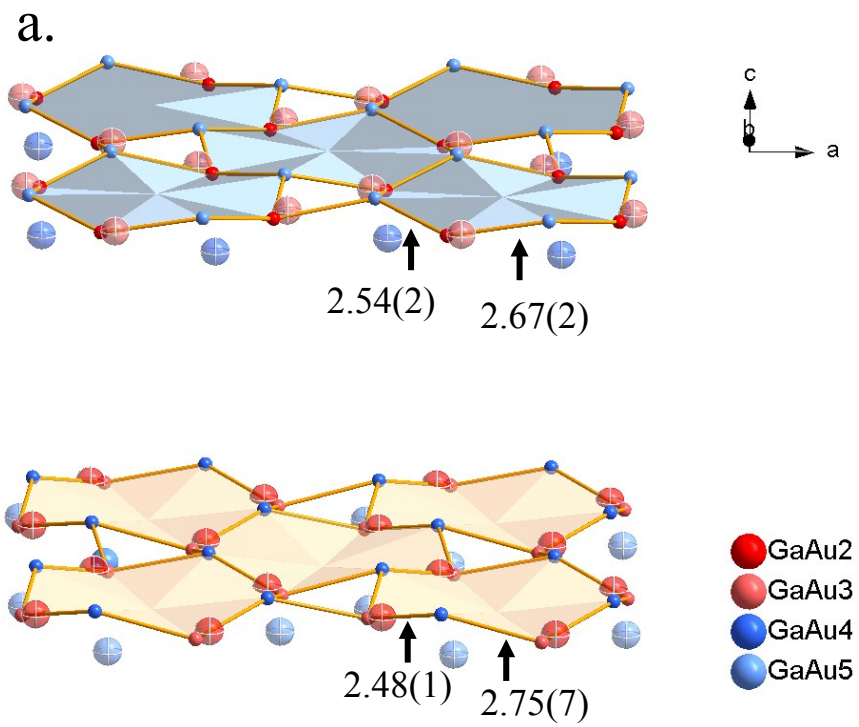


Figure S2. Polyhedral representation of $NaZn_{13}$ structure. Na-centered snub cubes are surrounded by Zn-filled icosahedra, and tetrahedral stars (outlined and pulled aside for clarity).



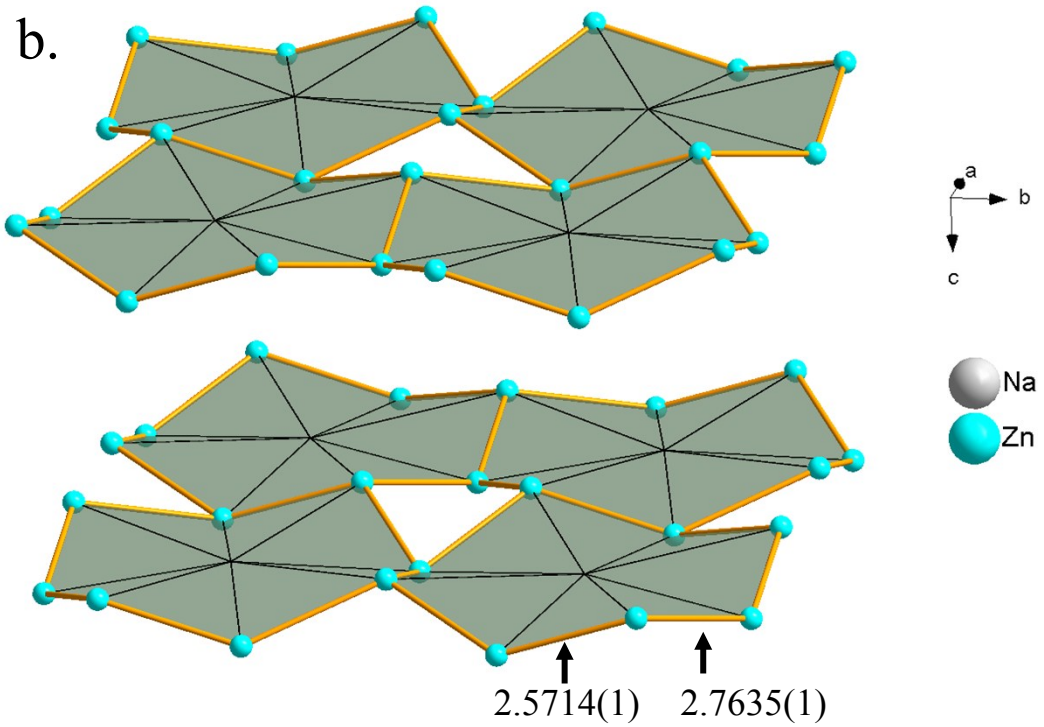


Figure S3. Octagonal nets in $BaAu_xGa_{12-x}$ (*a.*) and $NaZn_{13}$ (*b.*) with atomic distances labeled to show distortion within the layers. The disordered nets made up of (GaAu)2 and (GaAu)5 sites, and (GaAu)3 and (GaAu)4 sites are less and more distorted (respectively) than the nets in $NaZn_{13}$.