Electronic Supplementary Information (ESI)

Achieving Broadband Ultraviolet to Mid-Infrared Transparency in Germanate-based Nonlinear Optical Crystals $Cs_3REGe_3O_9$ (RE = Y, Gd)

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1. **Fig. S1** Powder XRD patterns of the $Cs_3YGe_3O_9$ and $Cs_3GdGe_3O_9$ polycrystalline powder before and after melting at 1250 °C, respectively.
2. **Fig. S2** EDS of $Cs_3YGe_3O_9$ (a) atomic ratio of various elements, (b) surface morphology, (c-f) Cs, Ge, Y, and O mapping results, respectively.
3. **Fig. S3** EDS of $Cs_3GdGe_3O_9$ (a) atomic ratio of various elements, (b) surface morphology, (c-f) Cs, Ge, Gd, and O mapping results, respectively.
4. **Table S1** Selected bond lengths (Å) and angles (deg.) for $Cs_3YGe_3O_9$.
5. **Table S2** Selected bond lengths (Å) and angles (deg.) for and $Cs_3GdGe_3O_9$.
6. **Table S3** The final atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) for $Cs_3YGe_3O_9$. U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor, and the bond valence sum for each atom in asymmetric unit.
7. **Table S4** The final atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) for $Cs_3GdGe_3O_9$. U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor, and the bond valence sum for each atom in asymmetric unit.
8. **Table S5** The detailed infrared peaks of CYGO and CGGO, respectively.

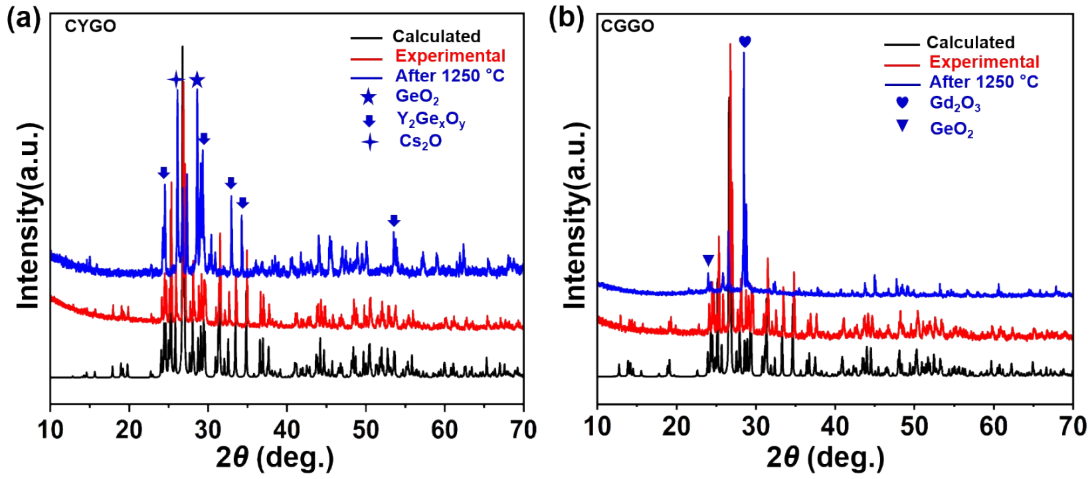

Fig. S1(a-b**)** Comparison of theoretical XRD spectra, experimentally obtained polycrystalline pure phase XRD spectra, and XRD spectra after calcination at 1250 °C for CYGO and CGGO, respectively.

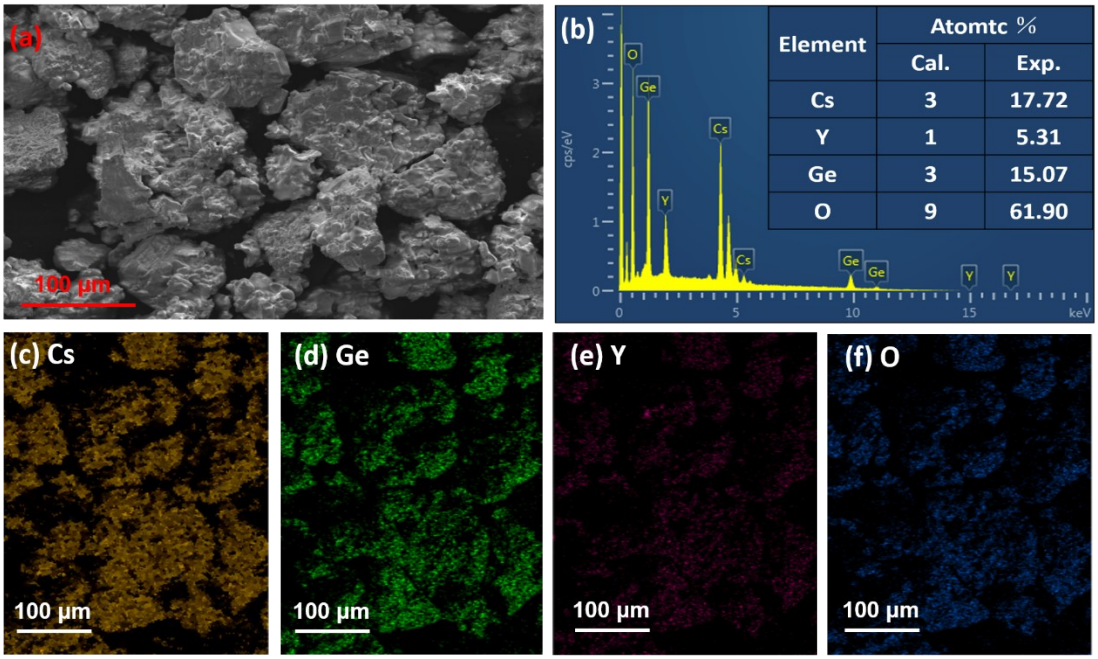

Fig. S2 EDS of $Cs_3YGe_3O_9$. (a) Atomic ratio of various elements, (b) Surface morphology, (c-f) Cs, Ge, Y, and O mapping results, respectively.

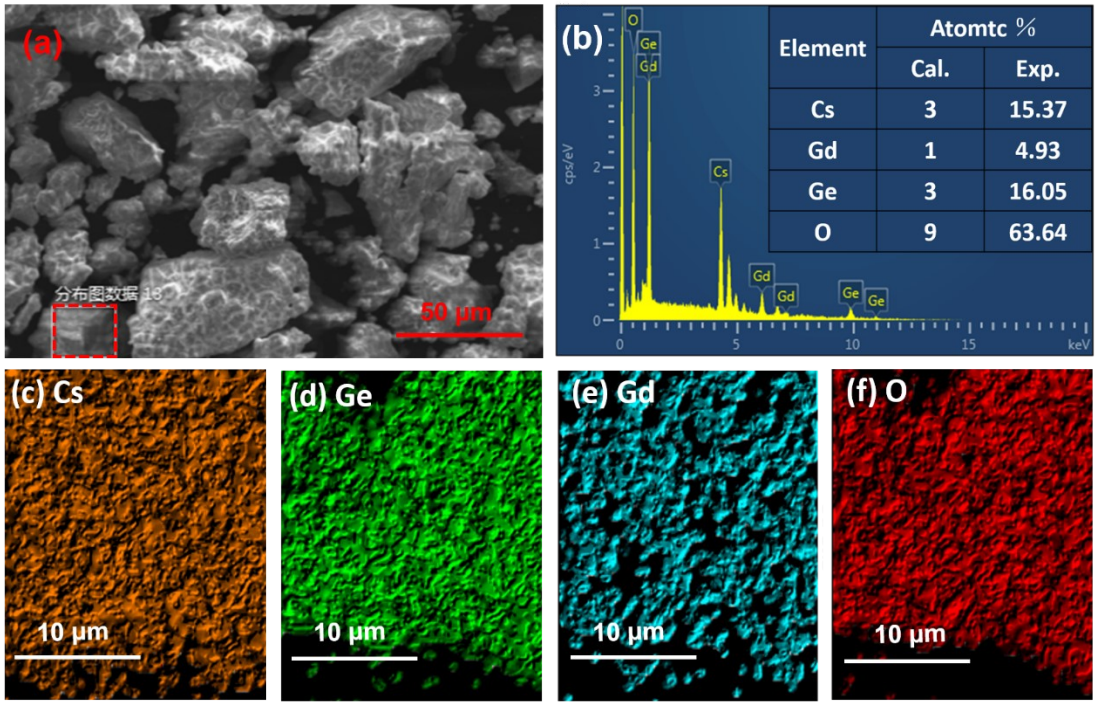

Fig. S3 EDS of $Cs_3GdGe_3O_9$. (a) Atomic ratio of various elements, (b) Surface morphology, (c-f) Cs, Ge, Gd, and O mapping results, respectively.

Table S1 Selected bond lengths (Å) and angles (deg.) for $Cs_3YGe_3O_9$.

Cs(1)-O(1)	2.993(6)	O(1)-Cs(3A)#9	3.004(19)
Cs(1)-O(5)#1	3.079(7)	O(4)-Cs(3A)#6	3.037(15)
Cs(1)-O(8)#2	3.098(7)	O(9)-Cs(3A)#9	3.064(16)
Cs(1)-O(4)#3	3.194(7)	O(3)-Cs(3A)#6	3.201(17)
Cs(1)-O(2)	3.381(7)	O(3)-Cs(3A)#9	3.53(3)
Cs(1)-O(9)#2	3.397(6)	O(7)-Cs(3A)	3.78(3)
Cs(2)-O(3)	3.041(6)	O(2)-Cs(3A)	2.855(17)
Cs(2)-O(8)#5	3.134(7)	Y(1)-O(8)#8	2.241(6)
Cs(2)-O(6)#2	3.146(7)	Y(1)-O(2)#3	2.244(7)
Cs(2)-O(5)#5	3.156(7)	Y(1)-O(3)#1	2.250(6)
Cs(2)-O(9)#2	3.230(7)	Y(1)-O(4)#3	2.259(6)
Cs(2)-O(7)#2	3.262(7)	Y(1)-O(1)	2.270(6)
Cs(3)-O(2)	2.925(9)	Y(1)-O(5)#1	2.281(6)
Cs(3)-O(4)#3	3.026(8)	Ge(1)-O(1)	1.721(6)
Cs(3)-O(1)#7	3.099(8)	Ge(1)-O(2)	1.722(7)
Cs(3)-O(9)#7	3.114(9)	Ge(1)-O(7)	1.786(6)
Cs(3)-O(3)#3	3.152(9)	Ge(1)-O(9)	1.789(6)
Cs(3)-O(7)	3.618(9)	Ge(3)-O(3)	1.712(6)
Cs(3)-O(3)#7	3.721(10)	Ge(3)-O(4)	1.723(6)
Ge(2)-O(8)	1.730(6)	Ge(3)-O(6)	1.763(6)
Ge(2)-O(5)	1.732(6)	Ge(3)-O(9)	1.774(6)
Ge(2)-O(7)#10	1.769(6)		
Ge(2)-O(6)	1.779(6)		
O(1)-Cs(1)-O(5)#1	61.80(15)	O(2)-Cs(3)-O(3)#7	56.22(19)
O(1)-Cs(1)-O(8)#2	103.52(15)	O(4)#3-Cs(3)-O(3)#7	127.1(2)
O(5)#1-Cs(1)-O(8)#2	157.69(18)	O(1)#7-Cs(3)-O(3)#7	81.4(2)
O(1)-Cs(1)-O(4)#3	60.96(16)	O(9)#7-Cs(3)-O(3)#7	47.97(17)
O(5)#1-Cs(1)-O(4)#3	60.90(17)	O(3)#3-Cs(3)-O(3)#7	138.58(17)

O(8)#2-Cs(1)-O(4)#3	129.15(17)	O(7)-Cs(3)-O(3)#7	99.51(18)
O(1)-Cs(1)-O(2)	53.96(15)	O(8)#8-Y(1)-O(2)#3	88.3(3)
O(5)#1-Cs(1)-O(2)	114.64(15)	O(8)#8-Y(1)-O(3)#1	86.7(2)
O(8)#2-Cs(1)-O(2)	57.48(15)	O(2)#3-Y(1)-O(3)#1	91.1(3)
O(4)#3-Cs(1)-O(2)	78.30(16)	O(8)#8-Y(1)-O(4)#3	97.6(2)
O(1)-Cs(1)-O(9)#2	114.69(16)	O(2)#3-Y(1)-O(4)#3	87.0(2)
O(5)#1-Cs(1)-O(9)#2	83.15(15)	O(3)#1-Y(1)-O(4)#3	175.3(2)
O(8)#2-Cs(1)-O(9)#2	89.02(15)	O(8)#8-Y(1)-O(1)	86.6(2)
O(4)#3-Cs(1)-O(9)#2	141.80(16)	O(2)#3-Y(1)-O(1)	172.2(2)
O(2)-Cs(1)-O(9)#2	132.69(16)	O(3)#1-Y(1)-O(1)	94.4(2)
O(3)-Cs(2)-O(8)#5	59.87(16)	O(4)#3-Y(1)-O(1)	87.9(2)
O(3)-Cs(2)-O(6)#2	115.89(17)	O(8)#8-Y(1)-O(5)#1	170.3(3)
O(8)#5-Cs(2)-O(6)#2	131.97(19)	O(2)#3-Y(1)-O(5)#1	99.2(3)
O(3)-Cs(2)-O(5)#5	103.39(16)	O(3)#1-Y(1)-O(5)#1	87.1(2)
O(8)#5-Cs(2)-O(5)#5	54.51(15)	O(4)#3-Y(1)-O(5)#1	88.9(2)
O(6)#2-Cs(2)-O(5)#5	85.26(19)	O(1)-Y(1)-O(5)#1	86.5(2)
O(3)-Cs(2)-O(9)#2	159.22(16)	O(1)-Ge(1)-O(2)	115.5(3)
O(8)#5-Cs(2)-O(9)#2	140.23(16)	O(1)-Ge(1)-O(7)	112.7(3)
O(6)#2-Cs(2)-O(9)#2	49.80(16)	O(2)-Ge(1)-O(7)	107.7(3)
O(5)#5-Cs(2)-O(9)#2	91.14(15)	O(1)-Ge(1)-O(9)	108.7(3)
O(3)-Cs(2)-O(7)#2	137.93(17)	O(2)-Ge(1)-O(9)	111.5(3)
O(8)#5-Cs(2)-O(7)#2	112.52(19)	O(7)-Ge(1)-O(9)	99.7(3)
O(6)#2-Cs(2)-O(7)#2	99.29(17)	O(8)-Ge(2)-O(5)	112.6(3)
O(5)#5-Cs(2)-O(7)#2	101.70(17)	O(8)-Ge(2)-O(7)#10	112.4(3)
O(9)#2-Cs(2)-O(7)#2	49.76(16)	O(5)-Ge(2)-O(7)#10	111.3(3)
O(2)-Cs(3)-O(4)#3	88.5(2)	O(8)-Ge(2)-O(6)	108.4(3)
O(2)-Cs(3)-O(1)#7	135.9(3)	O(5)-Ge(2)-O(6)	113.5(3)
O(4)#3-Cs(3)-O(1)#7	111.3(2)	O(7)#10-Ge(2)-O(6)	97.7(3)
O(2)-Cs(3)-O(9)#7	97.1(2)	O(3)-Ge(3)-O(4)	113.3(3)

O(4)#3-Cs(3)-O(9)#7	163.6(2)	O(3)-Ge(3)-O(6)	114.3(3)
O(1)#7-Cs(3)-O(9)#7	54.66(18)	O(4)-Ge(3)-O(6)	107.1(3)
O(2)-Cs(3)-O(3)#3	143.1(2)	O(3)-Ge(3)-O(9)	108.7(3)
O(4)#3-Cs(3)-O(3)#3	55.30(18)	O(4)-Ge(3)-O(9)	114.0(3)
O(1)#7-Cs(3)-O(3)#3	64.11(18)	O(6)-Ge(3)-O(9)	98.8(3)
O(9)#7-Cs(3)-O(3)#3	115.9(2)	O(1)#7-Cs(3)-O(7)	166.7(2)
O(2)-Cs(3)-O(7)	49.94(18)	O(9)#7-Cs(3)-O(7)	116.50(19)
O(4)#3-Cs(3)-O(7)	78.9(2)	O(3)#3-Cs(3)-O(7)	119.4(2)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,-y,z-1/2 #2 x+1/2,-y+1/2,z #3 -x+1,-y+1,z-1/2
#4 x+1/2,-y+3/2,z #5 x+1/2,-y-1/2,z #6 -x+1,-y+1,z+1/2
#7 x,y+1,z #8 -x+1/2,y+1/2,z-1/2 #9 x,y-1,z
#10 -x+1/2,y-1/2,z+1/2 #11 x-1/2,-y+3/2,z #12 -x+3/2,y+1/2,z-1/2
#13 x-1/2,-y+1/2,z #14 x-1/2,-y-1/2,z #15 -x+1,-y,z+1/2

Table S2 Selected bond lengths (Å) and angles (deg.) for $Cs_3GdGe_3O_9$.

Cs(1)-O(4)	2.91(2)	Cs(2A)-O(9)#10	2.69(3)
Cs(1)-O(2)#7	3.08(2)	Cs(2A)-O(6)#3	2.97(3)
Cs(1)-O(9)#8	3.13(2)	Cs(2A)-O(3)	2.97(3)
Cs(1)-O(3)#8	3.17(2)	Cs(2A)-O(8)	3.25(3)
Cs(1)-O(1)#7	3.23(2)	Cs(2A)-O(7)#3	3.27(4)
Cs(1)-O(5)	3.61(2)	Cs(2A)-O(4)#10	3.42(3)
Cs(1)-O(1)#8	3.69(2)	Cs(2A)-O(5)	3.49(3)
Cs(1A)-O(4)	2.86(3)	Gd(1)-O(4)#1	2.28(2)
Cs(1A)-O(9)#8	2.94(3)	Gd(1)-O(2)#1	2.286(19)
Cs(1A)-O(2)#7	3.03(3)	Gd(1)-O(1)	2.29(2)
Cs(1A)-O(3)#8	3.24(3)	Gd(1)-O(9)#2	2.31(2)
Cs(1A)-O(1)#7	3.30(3)	Gd(1)-O(8)#3	2.331(19)
Cs(1A)-O(1)#8	3.34(4)	Gd(1)-O(6)#2	2.33(2)
Cs(2)-O(9)#10	3.00(2)	Ge(1)-O(1)	1.71(2)
Cs(2)-O(6)#3	3.07(2)	Ge(1)-O(2)	1.73(2)

Cs(2)-O(8)	3.07(2)	Ge(1)-O(7)#3	1.75(2)
Cs(2)-O(2)#11	3.23(2)	Ge(1)-O(3)	1.78(2)
Cs(2)-O(4)#10	3.42(2)	Ge(2)-O(4)	1.73(2)
Cs(2)-O(3)	3.44(2)	Ge(2)-O(5)	1.75(2)
Cs(3)-O(1)#10	3.070(18)	Ge(2)-O(9)	1.76(2)
Cs(3)-O(8)#12	3.16(2)	Ge(2)-O(3)	1.769(19)
Cs(3)-O(6)#12	3.17(2)	Ge(3)-O(6)	1.71(2)
Cs(3)-O(7)#3	3.18(2)	Ge(3)-O(8)	1.746(19)
Cs(3)-O(3)	3.22(2)	Ge(3)-O(7)	1.80(2)
Cs(3)-O(5)	3.31(3)	Ge(3)-O(5)	1.82(2)
Cs(3A)-O(1)#10	2.70(3)	Cs(3A)-O(6)#12	3.03(4)
Cs(3A)-O(8)#12	2.92(4)	Cs(3A)-O(2)#10	3.75(4)
Cs(3A)-O(7)#3	2.98(4)	Cs(3A)-O(1)#10	2.70(3)
O(4)#1-Gd(1)-O(2)#1	86.9(8)	O(4)-Cs(1)-O(2)#7	89.0(6)
O(4)#1-Gd(1)-O(1)	90.8(8)	O(4)-Cs(1)-O(9)#8	137.3(6)
O(2)#1-Gd(1)-O(1)	174.4(7)	O(2)#7-Cs(1)-O(9)#8	110.1(6)
O(4)#1-Gd(1)-O(9)#2	173.2(8)	O(4)-Cs(1)-O(3)#8	98.9(6)
O(2)#1-Gd(1)-O(9)#2	88.3(8)	O(2)#7-Cs(1)-O(3)#8	163.1(6)
O(1)-Gd(1)-O(9)#2	94.4(8)	O(9)#8-Cs(1)-O(3)#8	54.5(5)
O(4)#1-Gd(1)-O(8)#3	99.7(8)	O(4)-Cs(1)-O(1)#7	142.8(6)
O(2)#1-Gd(1)-O(8)#3	87.7(7)	O(2)#7-Cs(1)-O(1)#7	54.2(5)
O(1)-Gd(1)-O(8)#3	87.7(7)	O(9)#8-Cs(1)-O(1)#7	64.0(5)
O(9)#2-Gd(1)-O(8)#3	84.8(8)	O(3)#8-Cs(1)-O(1)#7	115.2(5)
O(4)#1-Gd(1)-O(6)#2	88.3(8)	O(4)-Cs(1)-O(5)	49.0(5)
O(2)#1-Gd(1)-O(6)#2	97.4(8)	O(2)#7-Cs(1)-O(5)	79.2(5)
O(1)-Gd(1)-O(6)#2	87.6(7)	O(9)#8-Cs(1)-O(5)	167.0(6)
O(9)#2-Gd(1)-O(6)#2	87.6(8)	O(3)#8-Cs(1)-O(5)	117.2(5)
O(8)#3-Gd(1)-O(6)#2	170.8(8)	O(1)#7-Cs(1)-O(5)	118.7(6)
O(4)-Cs(1)-O(1)#8	57.6(6)	O(4)-Cs(1A)-O(9)#8	152.0(11)

O(2)#7-Cs(1)-O(1)#8	128.3(5)	O(4)-Cs(1A)-O(2)#7	90.8(8)
O(9)#8-Cs(1)-O(1)#8	81.6(5)	O(9)#8-Cs(1A)-O(2)#7	117.1(9)
O(3)#8-Cs(1)-O(1)#8	48.6(5)	O(4)-Cs(1A)-O(3)#8	98.2(9)
O(1)#7-Cs(1)-O(1)#8	139.0(3)	O(9)#8-Cs(1A)-O(3)#8	55.4(6)
O(5)-Cs(1)-O(1)#8	99.9(5)	O(2)#7-Cs(1A)-O(3)#8	160.7(16)
O(9)#10-Cs(2)-O(6)#3	104.2(6)	O(9)#8-Cs(1A)-O(1)#7	65.2(7)
O(9)#10-Cs(2)-O(8)	62.1(5)	O(2)#7-Cs(1A)-O(1)#7	53.9(6)
O(6)#3-Cs(2)-O(8)	157.6(6)	O(3)#8-Cs(1A)-O(1)#7	111.6(9)
O(9)#10-Cs(2)-O(2)#11	61.7(6)	O(4)-Cs(1A)-O(1)#8	62.7(8)
O(6)#3-Cs(2)-O(2)#11	130.7(6)	O(9)#8-Cs(1A)-O(1)#8	90.8(9)
O(8)-Cs(2)-O(2)#11	61.0(5)	O(2)#7-Cs(1A)-O(1)#8	146.5(11)
O(9)#10-Cs(2)-O(4)#10	53.9(5)	O(3)#8-Cs(1A)-O(1)#8	51.5(6)
O(6)#3-Cs(2)-O(4)#10	59.1(6)	O(1)#7-Cs(1A)-O(1)#8	155.7(8)
O(8)-Cs(2)-O(4)#10	114.7(5)	O(4)-Cs(1A)-O(1)#7	141.6(12)
O(2)#11-Cs(2)-O(4)#10	78.3(6)	O(1)#10-Cs(3)-O(8)#12	103.7(5)
O(9)#10-Cs(2)-O(3)	113.2(6)	O(1)#10-Cs(3)-O(6)#12	61.6(6)
O(6)#3-Cs(2)-O(3)	88.1(5)	O(8)#12-Cs(3)-O(6)#12	54.0(5)
O(8)-Cs(2)-O(3)	82.3(5)	O(1)#10-Cs(3)-O(7)#3	117.0(6)
O(2)#11-Cs(2)-O(3)	141.2(5)	O(8)#12-Cs(3)-O(7)#3	84.7(6)
O(4)#10-Cs(2)-O(3)	132.5(5)	O(6)#12-Cs(3)-O(7)#3	132.5(7)
O(9)#10-Cs(2A)-O(6)#3	115.7(9)	O(1)#10-Cs(3)-O(3)	159.9(5)
O(9)#10-Cs(2A)-O(3)	144.7(11)	O(8)#12-Cs(3)-O(3)	91.2(5)
O(6)#3-Cs(2A)-O(3)	99.6(9)	O(6)#12-Cs(3)-O(3)	138.2(5)
O(9)#10-Cs(2A)-O(8)	62.9(7)	O(7)#3-Cs(3)-O(3)	50.2(6)
O(6)#3-Cs(2A)-O(8)	151.0(12)	O(1)#10-Cs(3)-O(5)	137.2(6)
O(3)-Cs(2A)-O(8)	87.3(8)	O(8)#12-Cs(3)-O(5)	101.6(5)
O(9)#10-Cs(2A)-O(7)#3	155.1(11)	O(6)#12-Cs(3)-O(5)	110.2(6)
O(6)#3-Cs(2A)-O(7)#3	53.9(7)	O(7)#3-Cs(3)-O(5)	99.2(6)
O(3)-Cs(2A)-O(7)#3	51.3(7)	O(3)-Cs(3)-O(5)	49.2(5)

O(8)-Cs(2A)-O(7)#3	138.5(9)	O(1)#10-Cs(3A)-O(8)#12	121.2(13)
O(9)#10-Cs(2A)-O(4)#10	55.9(7)	O(1)#10-Cs(3A)-O(7)#3	139.2(15)
O(6)#3-Cs(2A)-O(4)#10	59.9(7)	O(8)#12-Cs(3A)-O(7)#3	92.7(11)
O(3)-Cs(2A)-O(4)#10	159.4(10)	O(1)#10-Cs(3A)-O(6)#12	67.5(8)
O(8)-Cs(2A)-O(4)#10	110.2(9)	O(8)#12-Cs(3A)-O(6)#12	57.7(9)
O(7)#3-Cs(2A)-O(4)#10	110.1(9)	O(7)#3-Cs(3A)-O(6)#12	150.4(14)
O(9)#10-Cs(2A)-O(5)	96.0(9)	O(1)#10-Cs(3A)-O(3)	146.7(16)
O(6)#3-Cs(2A)-O(5)	147.2(10)	O(8)#12-Cs(3A)-O(3)	85.3(9)
O(3)-Cs(2A)-O(5)	48.9(6)	O(7)#3-Cs(3A)-O(3)	45.8(7)
O(8)-Cs(2A)-O(5)	51.6(6)	O(6)#12-Cs(3A)-O(3)	123.0(12)
O(7)#3-Cs(2A)-O(5)	93.8(8)	O(1)#10-Cs(3A)-O(2)#10	49.8(7)
O(4)#10-Cs(2A)-O(5)	151.4(9)	O(8)#12-Cs(3A)-O(2)#10	146.1(15)
O(1)-Ge(1)-O(2)	113.7(11)	O(7)#3-Cs(3A)-O(2)#10	89.5(10)
O(1)-Ge(1)-O(7)#3	114.1(10)	O(6)#12-Cs(3A)-O(2)#10	114.9(9)
O(2)-Ge(1)-O(7)#3	105.2(12)	O(3)-Cs(3A)-O(2)#10	118.9(10)
O(1)-Ge(1)-O(3)	110.3(11)	O(6)-Ge(3)-O(8)	112.3(11)
O(2)-Ge(1)-O(3)	112.1(10)	O(6)-Ge(3)-O(7)	107.7(12)
O(7)#3-Ge(1)-O(3)	100.7(11)	O(8)-Ge(3)-O(7)	114.8(9)
O(4)-Ge(2)-O(5)	105.8(11)	O(6)-Ge(3)-O(5)	113.1(11)
O(4)-Ge(2)-O(9)	114.2(10)	O(8)-Ge(3)-O(5)	111.2(11)
O(5)-Ge(2)-O(9)	114.7(11)	O(7)-Ge(3)-O(5)	96.7(11)
O(4)-Ge(2)-O(3)	110.5(11)	O(5)-Ge(2)-O(3)	101.0(11)
O(9)-Ge(2)-O(3)	109.8(10)		

Symmetry transformations used to generate equivalent atoms:

#1 x,y+1,z #2 -x+1,-y+2,z+1/2 #3 -x+1/2,y+1/2,z+1/2
#4 x+1/2,-y+3/2,z #5 -x+1,-y+1,z+1/2 #6 -x+1/2,y-1/2,z-1/2
#7 -x+1,-y+1,z-1/2 #8 x,y-1,z #9 x+1/2,-y+1/2,z
#10 x-1/2,-y+3/2,z #11 -x+1/2,y+1/2,z-1/2 #12 -x+1/2,y-1/2,z+1/2
#13 x-1/2,-y+1/2,z #14 -x+1,-y+2,z-1/2

Table S3 The final atomic coordinates (× 10^4) and equivalent isotropic displacement parameters

($Å^2 \times 10^3$) for $Cs_3YGe_3O_9$. U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor, and the bond valence sum for each atom in asymmetric unit.

	x	y	z	U(eq)	BVS
Y(1)	5381(1)	3462(1)	2259(1)	7(1)	3.107
Ge(1)	4114(1)	3958(1)	4781(1)	9(1)	3.947
Ge(2)	2244(1)	-1076(1)	7751(1)	10(1)	3.961
Ge(3)	3953(1)	1544(1)	6882(1)	8(1)	4.063
Cs(1)	6719(1)	5445(1)	4680(1)	32(1)	0.810
Cs(2)	6774(1)	501(1)	6220(1)	34(1)	0.824
Cs(3)	4370(3)	8944(7)	4025(5)	32(1)	0.970
Cs(3A)	4540(20)	9010(20)	4160(20)	59(3)	1.076
O(1)	5028(4)	3112(9)	3991(5)	14(1)	-2.154
O(2)	4414(5)	5866(10)	5549(5)	22(2)	-2.249
O(3)	4709(4)	-366(8)	6901(6)	15(1)	-2.155
O(4)	4401(5)	3440(9)	7573(5)	18(1)	-2.092
O(5)	3042(4)	-2955(9)	7716(6)	19(1)	-1.836
O(6)	2778(5)	1095(9)	7365(6)	26(2)	-2.020
O(7)	3047(5)	4551(11)	4059(5)	24(2)	-2.015
O(8)	1224(4)	1451(10)	6989(5)	20(1)	-1.895
O(9)	3652(4)	2062(9)	5558(5)	16(1)	-2.340

Table S4 The final atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) for $Cs_3GdGe_3O_9$. U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor, and the bond valence sum for each atom in asymmetric unit.

	x	y	z	U(eq)	BVS
Gd(1)	4615(1)	13471(2)	7453(1)	11(1)	3.150
Ge(1)	3947(2)	8468(4)	7073(2)	14(1)	4.068
Ge(2)	4101(2)	6054(4)	4978(2)	13(1)	3.947
Ge(3)	2750(2)	6079(4)	2956(3)	16(1)	3.786
Cs(1)	4378(6)	1066(5)	4259(4)	55(2)	0.895
Cs(1A)	4900(30)	950(30)	4520(20)	55(2)	1.025
Cs(2)	1720(2)	10453(4)	4889(2)	43(1)	0.787
Cs(2A)	1591(15)	8800(40)	5410(20)	43(1)	1.272
Cs(3)	1774(2)	5513(5)	6445(3)	55(1)	0.771
Cs(3A)	1580(20)	5580(40)	7150(30)	55(1)	1.187
O(1)	4695(12)	10350(30)	7089(18)	24(5)	-1.980

O(2)	4383(16)	6570(30)	7766(17)	22(5)	-1.870
O(3)	3643(14)	7870(30)	5768(17)	23(4)	-2.172
O(4)	4398(15)	4140(30)	5735(19)	26(5)	-1.944
O(5)	3069(16)	5410(30)	4280(20)	33(6)	-1.940
O(6)	3730(15)	6400(30)	2168(18)	31(6)	-1.890
O(7)	2196(15)	3900(30)	2590(20)	41(7)	-1.980
O(8)	1979(13)	8000(30)	2950(18)	22(4)	-1.800
O(9)	5050(15)	6900(30)	4205(16)	23(5)	-1.840

Table S5 The infrared feature peak details of CYGO and CGGO.[1,2]

Compound	Position of peak	Vibration mode
CYGO and CGGO	400 cm^{-1}	Symmetrical curvature vibration of Ge-O-Ge
	532 cm^{-1}	Aymmetrical stretching vibrations of Ge-O
	731 cm^{-1}	Asymmetric stretching of Ge-O-Ge

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