Supplementary Information

A Congruently Melting and Deep UV Nonlinear Optical Material: $Li_3Cs_2B_5O_{10}$

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Figure S1. Experimental and calculated XRD patterns of the $Li_3Cs_2B_5O_{10}$.

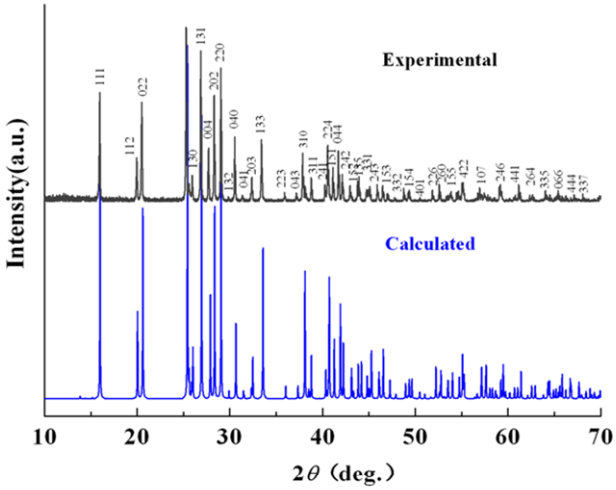

Figure S2. Powder XRD pattern of the $Li_3Cs_2B_5O_{10}$ samples before and after melting, respectively.

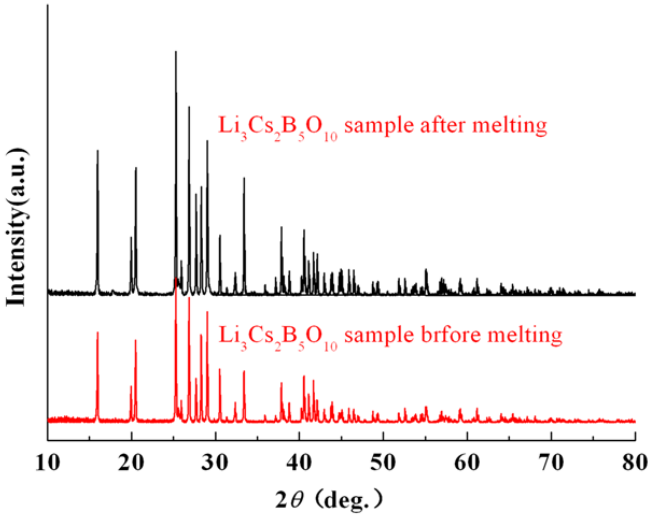

Figure S3. $Cs(1)O_{10}$ and $Cs(2)O_8$ polyhedra are interconnected via sharing oxygen atoms into a 3D framework. (purple, Cs; red, O).

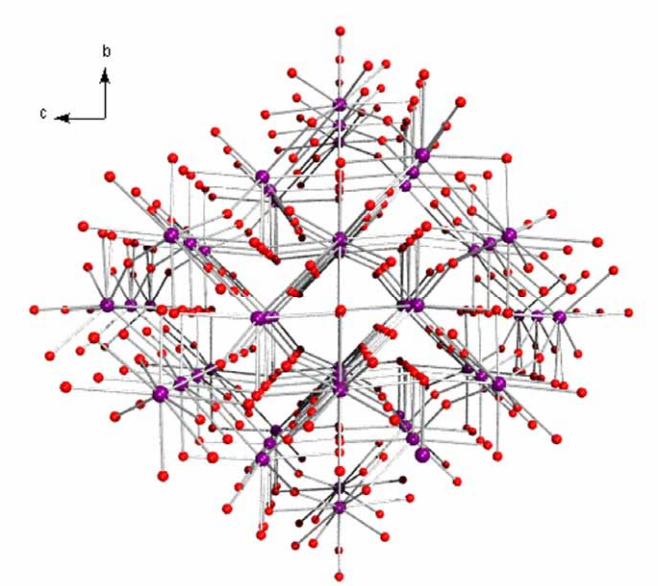

Figure S4 $Li(1)O_5$ and $Li(2)O_4$ polyhedra are interconnected via sharing edges into a 2D sheet network. (green, Li; red, O)

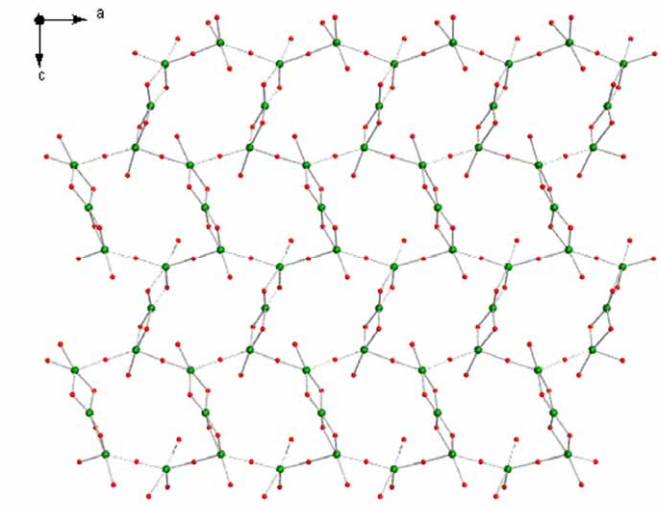

Figure S5 Phase-matching curve, that is, particle size vs. SHG intensity for $Li_3Cs_2B_5O_{10}$. The curve drawn is a guide to the eye and is not a fit to the data.

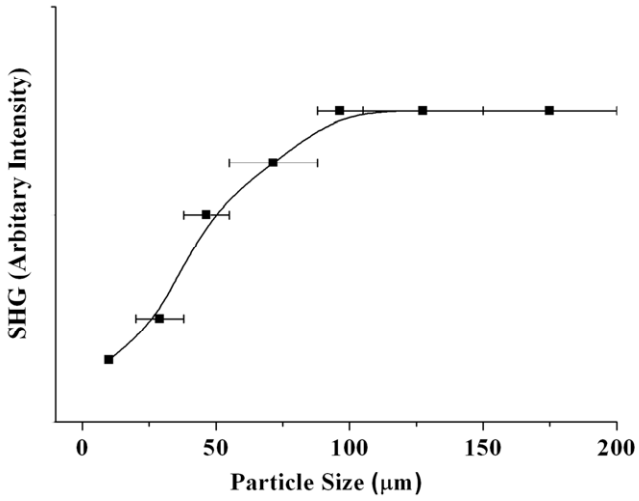

Table S1. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) for $Li_3Cs_2B_5O_{10}$. U_{Eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor.

Atom	x	y	z	U_{eq}
Cs(1)	2038(1)	5000	5000	15(1)
Cs(2)	0	2275(1)	2500	17(1)
O(1)	1153(4)	7555(2)	5015(3)	13(1)
O(2)	-2249(5)	5967(2)	5882(2)	14(1)
O(3)	-3357(4)	4749(2)	2496(3)	17(1)
O(4)	-64(6)	8249(2)	3419(2)	17(1)
O(5)	1196(4)	5830(2)	1522(2)	13(1)
B(1)	-5000	4012(5)	2500	15(1)
B(2)	2243(6)	4915(4)	1641(3)	10(1)
B(3)	1261(6)	8259(4)	4209(3)	11(1)
Li(1)	1312(11)	6918(7)	450(6)	18(2)
Li(2)	0	6723(8)	2500	12(2)

Table S2. Selected bond distances (Å) and angles (deg) for $Li_3Cs_2B_5O_{10}$.

Cs(1)-O(1)	3.047(3)	B(1)-O(3)#14	1.465(4)
Cs(1)-O(1)#1	3.047(3)	B(1)-O(4)#8	1.474(4)
Cs(1)-O(5)#2	3.191(3)	B(1)-O(4)#6	1.474(4)
Cs(1)-O(5)#3	3.191(3)	B(1)-O(3)	1.465(4)
Cs(1)-O(3)#3	3.343(4)	B(2)-O(3)#2	1.380(6)
Cs(1)-O(3)#2	3.343(4)	B(2)-O(2)#7	1.413(5)
Cs(1)-O(2)#1	3.484(3)	B(2) -O(5)	1.316(6)
Cs(1)-O(2)	3.484(3)	B(3)-O(2)#4	1.410(5)
Cs(1)-O(4)#4	3.554(4)	B(3)-O(1)	1.319(6)
Cs(1)-O(4)#5	3.554(4)	B(3) -O(4)	1.391(5)
Cs(2)-O(3)#6	3.176(2)	Li(1)-O(1)#15	2.010(9)
Cs(2)-O(3)#5	3.176(2)	Li(1)-O(1)#2	2.020(9)
Cs(2)-O(1)#1	3.289(4)	Li(1)-O(2)#2	2.141(8)
Cs(2)-O(1)#7	3.289(4)	Li(1)-O(4)#2	2.305(8)
Cs(2)-O(2)#1	3.334(3)	Li(1)-O(5)	1.869(8)
Cs(2)-O(2)#7	3.334(3)	Li(2)-O(5)#2	1.842(6)
Cs(2)-O(5)#8	3.457(3)	Li(2)-O(4)	2.133(8)

Cs(2)-O(5)#9	3.457(3)	Li(2)-O(4)#2	2.133(8)
O(1)-Cs(1)-O(1)#1	155.75(10)	Li(2)-O(5)	1.842(6)
O(1)-Cs(1)-O(5)#2	63.48(8)	O(3)#6-Cs(2)-O(3)#5	43.89(10)
O(1)#1-Cs(1)-O(5)#2	97.98(8)	O(3)#6-Cs(2)-O(1)#1	98.56(9)
O(1)-Cs(1)-O(5)#3	97.98(8)	O(3)#5-Cs(2)-O(1)#1	87.86(9)
O(1)#1-Cs(1)-O(5)#3	63.48(8)	O(3)#6-Cs(2)-O(1)#7	87.86(9)
O(5)#2-Cs(1)-O(5)#3	85.82(11)	O(3)#5-Cs(2)-O(1)#7	98.56(9)
O(1)-Cs(1)-O(3)#3	88.17(8)	O(1)#1-Cs(2)-O(1)#7	173.11(9)
O(1)#1-Cs(1)-O(3)#3	98.73(9)	O(3)#6-Cs(2)-O(2)#1	112.77(8)
O(5)#2-Cs(1)-O(3)#3	139.83(7)	O(3)#5-Cs(2)-O(2)#1	138.87(9)
O(5)#3-Cs(1)-O(3)#3	69.73(7)	O(1)#1-Cs(2)-O(2)#1	59.18(7)
O(1)-Cs(1)-O(3)#2	98.73(9)	O(1)#7-Cs(2)-O(2)#1	116.00(7)
O(1)#1-Cs(1)-O(3)#2	88.17(8)	O(3)#6-Cs(2)-O(2)#7	138.87(9)
O(5)#2-Cs(1)-O(3)#2	69.73(7)	O(3)#5-Cs(2)-O(2)#7	112.77(8)
O(5)#3-Cs(1)-O(3)#2	139.83(7)	O(1)#1-Cs(2)-O(2)#7	116.00(7)
O(3)#3-Cs(1)-O(3)#2	146.86(10)	O(1)#7-Cs(2)-O(2)#7	59.18(7)
O(1)-Cs(1)-O(2)#1	97.56(7)	O(2)#1-Cs(2)-O(2)#7	104.15(10)
O(1)#1-Cs(1)-O(2)#1	59.66(7)	O(3)#6-Cs(2)-O(5)#8	41.54(7)

O(5)#2-Cs(1)-O(2)#1	41.38(7)	O(3)#5-Cs(2)-O(5)#8	81.06(7)
O(5)#3-Cs(1)-O(2)#1	56.49(7)	O(1)#1-Cs(2)-O(5)#8	125.43(7)
O(3)#3-Cs(1)-O(2)#1	126.21(7)	O(1)#7-Cs(2)-O(5)#8	58.59(7)
O(3)#2-Cs(1)-O(2)#1	85.20(7)	O(2)#1-Cs(2)-O(5)#8	97.83(7)
O(1)-Cs(1)-O(2)	59.66(7)	O(2)#7-Cs(2)-O(5)#8	117.56(7)
O(1)#1-Cs(1)-O(2)	97.56(7)	O(3)#6-Cs(2)-O(5)#9	81.06(7)
O(5)#2-Cs(1)-O(2)	56.49(7)	O(3)#5-Cs(2)-O(5)#9	41.54(7)
O(5)#3-Cs(1)-O(2)	41.38(7)	O(1)#1-Cs(2)-O(5)#9	58.59(7)
O(3)#3-Cs(1)-O(2)	85.20(7)	O(1)#7-Cs(2)-O(5)#9	125.43(7)
O(3)#2-Cs(1)-O(2)	126.21(7)	O(2)#1-Cs(2)-O(5)#9	117.56(7)
O(2)#1-Cs(1)-O(2)	54.46(10)	O(2)#7-Cs(2)-O(5)#9	97.83(7)
O(1)-Cs(1)-O(4)#4	63.82(8)	O(5)#8-Cs(2)-O(5)#9	121.68(9)
O(1)#1-Cs(1)-O(4)#4	133.53(8)	O(3)-B(1)-O(3)#14	108.2(4)
O(5)#2-Cs(1)-O(4)#4	127.15(7)	O(3)-B(1)-O(4)#8	112.4(2)
O(5)#3-Cs(1)-O(4)#4	105.02(7)	O(3)#14-B(1)-O(4)#8	109.0(2)
O(3)#3-Cs(1)-O(4)#4	40.45(6)	O(3)-B(1)-O(4)#6	109.0(2)
O(3)#2-Cs(1)-O(4)#4	115.15(7)	O(3)#14-B(1)-O(4)#6	112.4(2)
O(2)#1-Cs(1)-O(4)#4	153.33(8)	O(4)#8-B(1)-O(4)#6	105.8(4)

O(2)-Cs(1)-O(4)#4	98.87(8)	O(5)-B(2)-O(3)#2	122.8(4)
O(1)-Cs(1)-O(4)#5	133.53(8)	O(5)-B(2)-O(2)#7	120.8(4)
O(1)#1-Cs(1)-O(4)#5	63.82(8)	O(3)#2-B(2)-O(2)#7	116.4(4)
O(5)#2-Cs(1)-O(4)#5	105.02(7)	O(1)-B(3)-O(4)	121.4(4)
O(5)#3-Cs(1)-O(4)#5	127.15(7)	O(1)-B(3)-O(2)#4	120.5(4)
O(3)#3-Cs(1)-O(4)#5	115.15(7)	O(4)-B(3)-O(2)#4	118.0(4)
O(3)#2-Cs(1)-O(4)#5	40.45(6)	O(5)-Li(1)-O(4)#2	88.8(3)
O(2)#1-Cs(1)-O(4)#5	98.87(8)	O(1)#15-Li(1)-O(4)#2	108.9(4)
O(2)-Cs(1)-O(4)#5	153.33(8)	O(1)#2-Li(1)-O(4)#2	65.9(3)
O(4)#4-Cs(1)-O(4)#5	107.80(11)	O(2)#2-Li(1)-O(4)#2	166.1(4)
O(5)-Li(1)-O(1)#15	116.8(4)	O(5)-Li(2)-O(5)#2	111.2(5)
O(5)-Li(1)-O(1)#2	115.2(4)	O(5)-Li(2)-O(4)	148.8(3)
O(1)#15-Li(1)-O(1)#2	127.6(4)	O(5)#2-Li(2)-O(4)	95.03(16)
O(5)-Li(1)-O(2)#2	104.2(4)	O(5)-Li(2)-O(4)#2	95.03(16)
O(1)#15-Li(1)-O(2)#2	69.5(3)	O(5)#2-Li(2)-O(4)#2	148.8(3)
O(1)#2-Li(1)-O(2)#2	103.6(4)	O(4)-Li(2)-O(4)#2	66.9(3)

Note. Symmetry transformations used to generate equivalent atoms:

#1 x,-y+1,-z+1	#2 -x,y,-z+1/2	#3 -x,-y+1,z+1/2
#4 x+1/2,-y+3/2,-z+1	#5 x+1/2,y-1/2,z	#6 -x-1/2,y-1/2,-z+1/2

#7 -x,-y+1,z-1/2	#8 x-1/2,y-1/2,z	#9 -x+1/2,y-1/2,-z+1/2
#10 -x+1/2,-y+3/2,z+1/2	#11 x-1/2,-y+3/2,-z+1	#12 x-1/2,y+1/2,z
#13 x+1/2,y+1/2,z	#14 -x-1,y,-z+1/2	#15 -x+1/2,-y+3/2,z-1/2

Table S3. Bond valence analysis of the $Li_3Cs_2B_5O_{10}$.[a,b]

Atoms	l	s	Atoms	l	s
Cs(1)-O(1)	3.045(3)	0.183	Cs(2)-O(3)#6	3.178(3)	0.128
Cs(1)-O(1)#1	3.045(3)	0.183	Cs(2)-O(3)#5	3.178(3)	0.128
Cs(1)-O(5)#2	3.187(4)	0.125	Cs(2)-O(1)#7	3.289(5)	0.095
Cs(1)-O(5)#3	3.187(4)	0.125	Cs(2)-O(1)#1	3.289(5)	0.095
Cs(1)-O(3)#3	3.340(5)	0.083	Cs(2)-O(2)#7	3.326(4)	0.109
Cs(1)-O(3)#2	3.340(5)	0.083	Cs(2)-O(2)#1	3.326(4)	0.109
Cs(1)-O(2)#1	3.478(4)	0.057	Cs(2)-O(5)#8	3.462(4)	0.059
Cs(1)-O(2)	3.478(4)	0.057	Cs(2)-O(5)#9	3.462(4)	0.059
Cs(1)-O(4)#4	3.558(4)	0.046	Σ s		0.782
Cs(1)-O(4)#5	3.558(4)	0.046			
Σ s		0.988			
Li(1)-O(1)#15	2.017(11)	0.226	Li(2)-O(4)	2.139(10)	0.162
Li(1)-O(1)#2	2.017(11)	0.226	Li(2)-O(5)	1.834(7)	0.370
Li(1)-O(2)#2	2.139(10)	0.162	Li(2)-O(5)#2	1.834(7)	0.370
Li(1)-O(4)#2	2.307(10)	0.103	Li(2)-O(4)#2	2.139(10)	0.162
Li(1)-O(5)	1.863(11)	0.342	Σ s		1.064
Σ s		1.059			

B(1)-O(3)	1.468(5)	0.769	B(2)-O(5)	1.320(7)	1.148
B(1)-O(3)#14	1.468(5)	0.769	B(2)-O(3)#2	1.381(7)	0.973
B(1)-O(4)#6	1.472(5)	0.761	B(2)-O(2)#7	1.414(6)	0.890
B(1)-O(4)#8	1.472(5)	0.761	Σ s		3.011
Σ s		3.060			

[a] Bond valences calculated with the program Bond Valence Calculator Version 2.00, Hormillosa, C., Healy, S., Stephen, T. McMaster University (1993).

Valence sums calculated with the formula: $S_i = \exp[(R_0-R_i)/B]$, where S_i = valence of bond "i" and $B = 0.37$. Superscripts indicate the number of equivalent bonds for anions.