Supporting Information

$Ba_{13}In_{12}Zn_7S_{38}$ and $Ba_{12}In_{12}Zn_8Se_{38}$: Infrared Nonlinear Optical Chalcogenides Designed by Zinc-Induced Non-Centrosymmetry Transformation in Ba-In-Q (Q = S, Se) System

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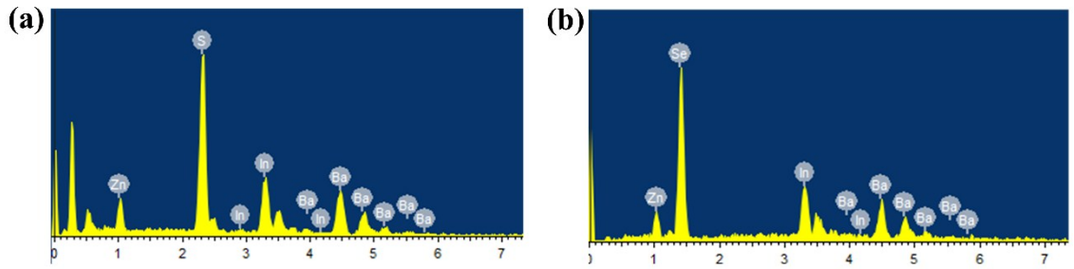

Figure S1. The EDX spectra of **1 (a)** and **2 (b)**.

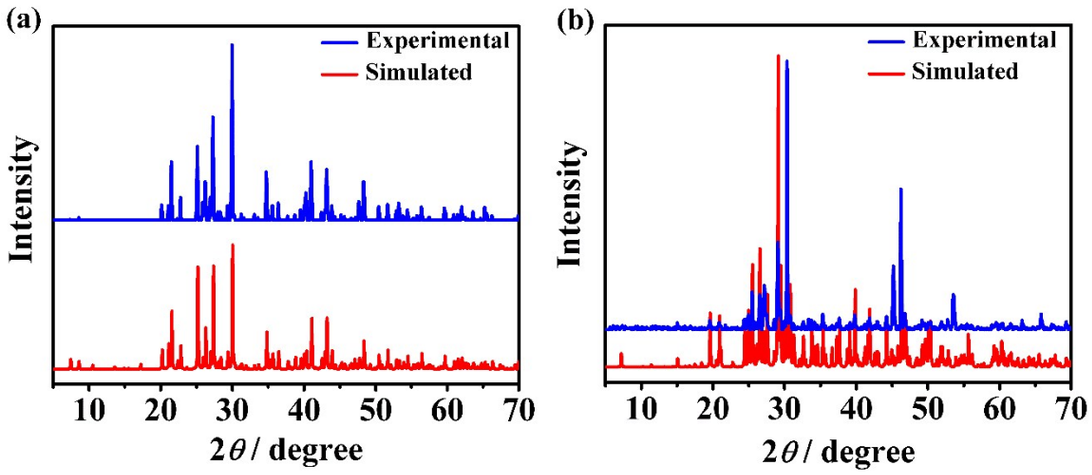

Figure S2. Experimental and simulated powder XRD patterns of **1 (a)** and **2 (b)**. Intensities of some peaks on the experimental patterns don't match those on the simulated ones because of the effect of preferred orientation.

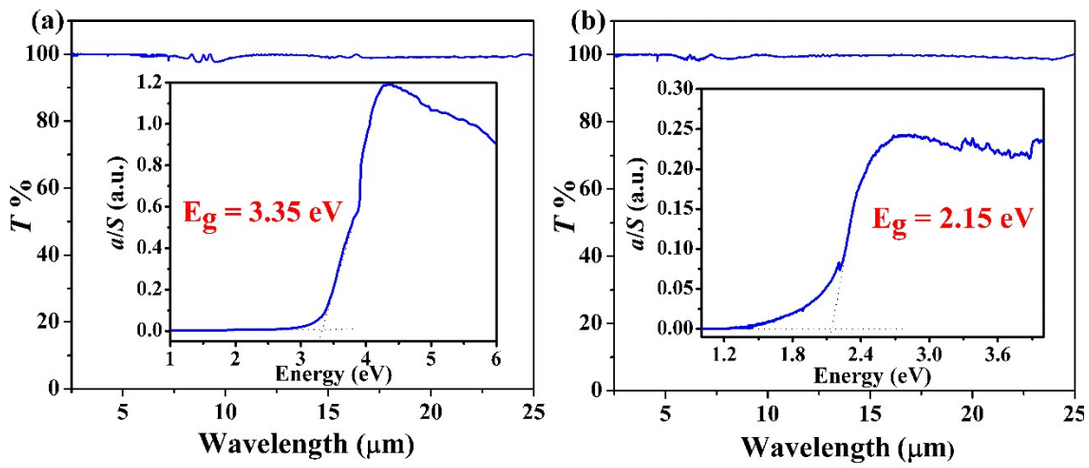

Figure S3. IR and UV-vis spectra of **1 (a)** and **2 (b)**.

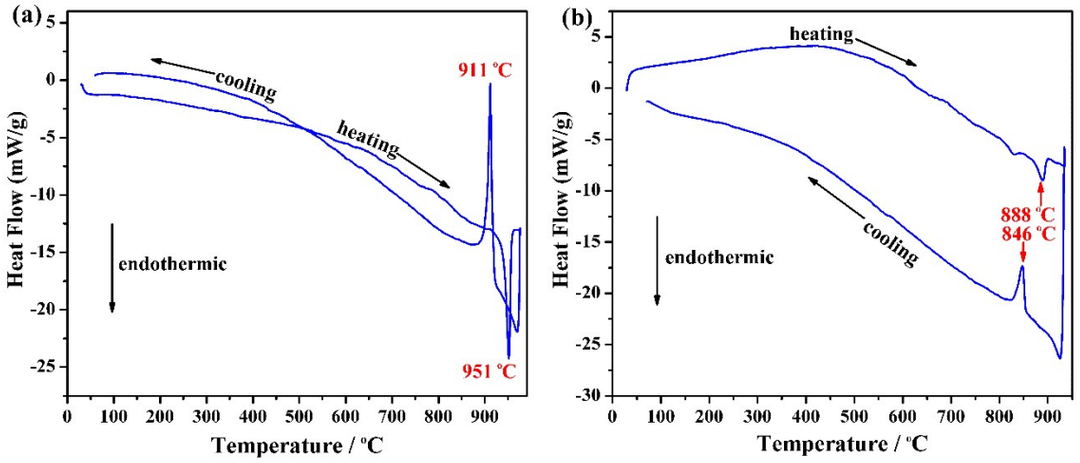

Figure S4. The DSC curves of **1 (a)** and **2 (b)**.

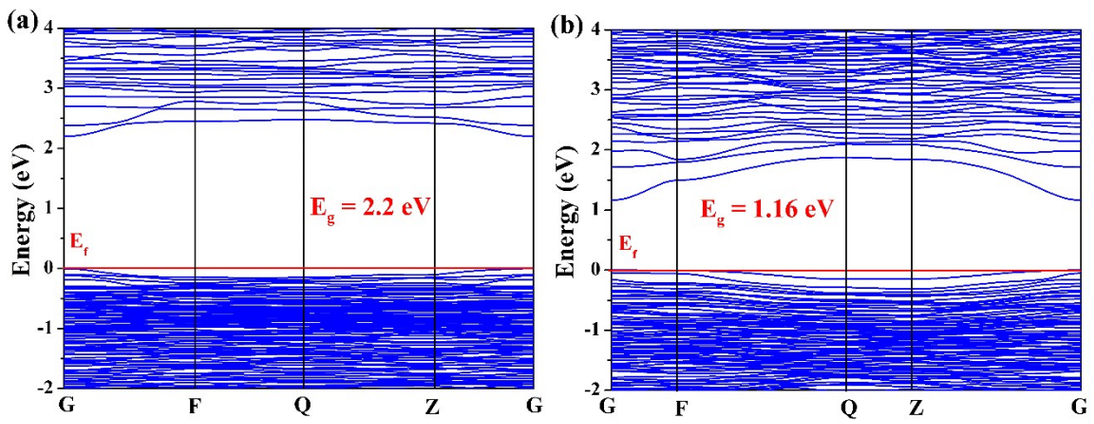

Figure S5. Calculated band structures of **1 (a)** and **2 (b)**. Fermi level is set at 0 eV for both compounds.

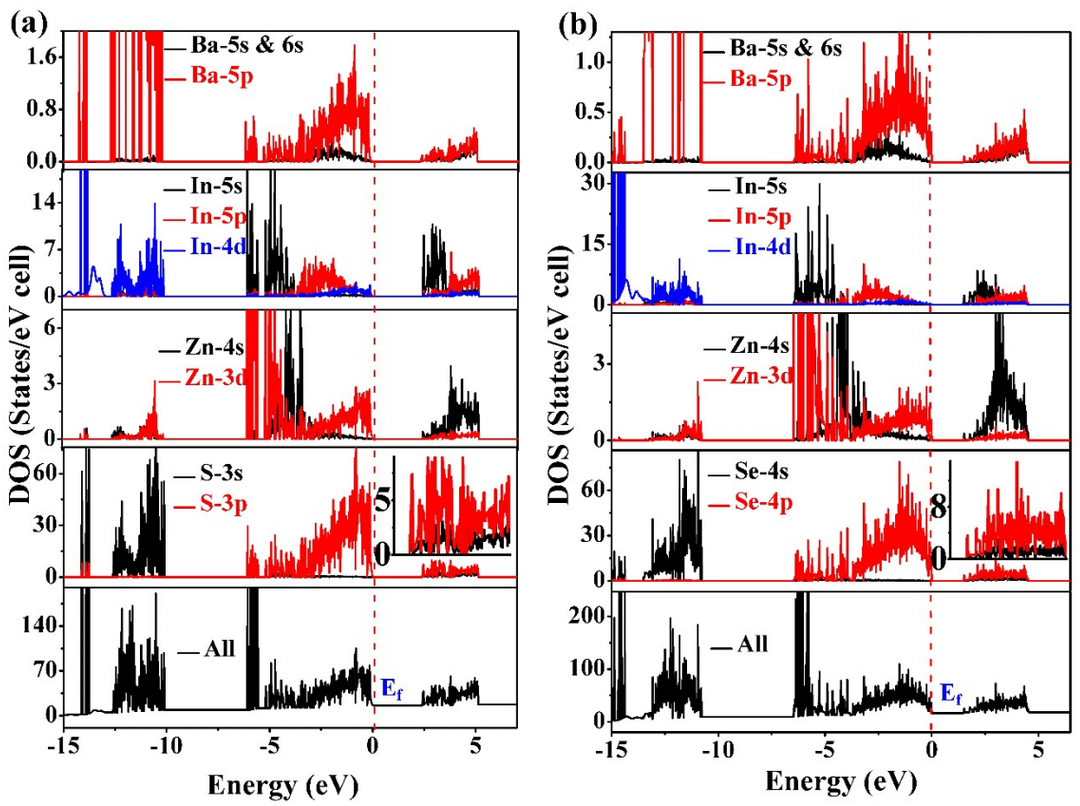

Figure S6. Total and partial densities of states of **1 (a)** and **2 (b).** Fermi level is set at 0

eV for both compounds.

Table S1. Atom coordinates and equivalent isotropic displacement parameters of **1** and **2**.

Atom	Wyckoff site	*x*	*y*	*z*	Occu.	***U*eq(Å^2)**
			1			
Ba1	8i	0.35409(2)	0.64591(2)	0.23061(2)		0.01272(6)
Ba2	8f	0.72263(2)	1.00000	0		0.01421(6)
Ba3	8g	0.72293(2)	1.00000	1/2		0.01434(6)
Ba4	2a	1/2	1/2	1/2		0.03229(18)
In1	4d	1/2	1.00000	1/4		0.01252(9)
In2	8i	0.72708(2)	0.72708(2)	0.48246(2)		0.01208(7)
In3	8i	0.60633(2)	0.60633(2)	0.19124(3)		0.01989(8)
In4	2b	1/2	1/2	0	0.505	0.505(7)
Zn1	16j	0.62612(2)	0.81503(2)	0.23489(3)	0.875	0.01285(8)
In5	16j	0.62612(2)	0.81503(2)	0.23489(3)	0.125	0.01285(8)
S1	16j	0.67375(6)	0.84368(5)	0.39767(6)		0.01302(15)
S2	8i	0.60700(5)	0.60700(5)	0.00071(9)		0.01187(19)
S3	6j	0.52573(5)	0.71787(5)	0.24963(7)		0.01296(14)
S4	8i	0.73320(5)	0.73320(5)	0.15878(8)		0.0117(2)
S5	16j	0.58540(5)	0.92012(5)	0.13456(6)		0.01357(16)
S6	8i	0.62778(5)	0.62778(5)	0.52561(9)		0.0138(2)
S7	4e	1/2	1/2	0.2336(2)		0.0274(4)
In44	4e	1/2	1/2	0.0232(5)	0.247	0.0031(14)
			2			
Ba1	4e	0.63671(2)	0.13671(2)	1.08628(9)		0.01228(13)
Ba2	8f	0.49227(2)	0.27301(3)	-0.46064(7)		0.01501(10)
In1	2a	1/2	1/2	0		0.0130(2)
In2	4e	0.89895(3)	0.39895(3)	0.08043(13)	0.729	0.01472(19)
Zn6	4e	0.89895(3)	0.39895(3)	0.08043(13)	0.271	0.01472(19)
In3	8f	0.68313(3)	0.36044(3)	-0.04912(10)	0.5596	0.01238(13)
Zn7	8f	0.68313(3)	0.36044(3)	-0.04912(10)	0.4404	0.01238(13)
Zn1	4e	0.77213(4)	0.27213(4)	0.43025(17)	0.848	0.0140(2)
In5	4e	0.77213(4)	0.27213(4)	0.43025(17)	0.152	0.0140(2)
In4	2c	1.00000	1/2	-0.4611(10)	0.35	0.0130(15)
Se1	2c	1.00000	1/2	-0.0577(2)		0.0104(2)
Se2	4e	0.87542(4)	0.37542(4)	0.45194(17)		0.01072(18)
Se3	8f	0.57839(4)	0.41984(4)	-0.25305(11)		0.01249(14)
Se4	8f	0.78043(4)	0.46914(4)	-0.04042(12)		0.01353(14)
Se5	8f	0.65060(4)	0.32020(4)	0.29613(11)		0.01207(14)
Se6	4e	0.90099(4)	0.59901(4)	-0.56496(16)		0.01155(19)
Se7	4e	0.74032(4)	0.24032(4)	0.78193(17)		0.0140(2)
In44	4e	1.0128(3)	0.5128(3)	-0.4626(7)	0.325	0.0138(15)

Table S2. Selected bond lengths (Å) of **1** and **2**.

1		**2**	
Bond	Distance	Bond	Distance
Ba(1)-S(3)	3.1403(8)	Ba(1)-Se(4)	3.2552(7)
Ba(1)-S(3)	3.1404(8)	Ba(1)-Se(4)	3.2552(7)
Ba(1)-S(2)	3.1514(12)	Ba(1)-Se(7)	3.2565(12)
Ba(1)-S(6)	3.2038(12)	Ba(1)-Se(6)	3.3353(12)
Ba(1)-S(4)	3.2164(12)	Ba(1)-Se(1)	3.3570(6)
Ba(1)-S(1)	3.4674(10)	Ba(1)-Se(5)	3.4859(9)
Ba(1)-S(1)	3.4674(10)	Ba(1)-Se(5)	3.4859(9)
Ba(1)-S(7)	3.4712(3)	Ba(1)-Se(6)	3.6105(12)
Ba(2)-S(5)	3.1944(9)	Ba(2)-Se(3)	3.2619(8)
Ba(2)-S(5)	3.1944(9)	Ba(2)-Se(2)	3.2753(5)
Ba(2)-S(6)	3.3261(3)	Ba(2)-Se(5)	3.2947(8)
Ba(2)-S(6)	3.3261(3)	Ba(2)-Se(5)	3.4444(9)
Ba(2)-S(1)	3.4004(9)	Ba(2)-Se(3)	3.4478(9)
Ba(2)-S(1)	3.4004(9)	Ba(2)-Se(4)	3.4896(10)
Ba(2)-S(3)	3.4379(9)	Ba(2)-Se(4)	3.5031(10)
Ba(2)-S(3)	3.4379(9)	Ba(2)-Se(6)	3.5429(5)
Ba(3)-S(1)	3.0617(8)	In(1)-Se(3)	2.5783(7)
Ba(3)-S(1)	3.0617(9)	In(1)-Se(3)	2.5783(7)
Ba(3)-S(5)	3.3052(9)	In(1)-Se(3)	2.5783(7)
Ba(3)-S(5)	3.3052(9)	In(1)-Se(3)	2.5784(7)
Ba(3)-S(2)	3.3798(4)	In(1)/Zn(5)-Se(5)	2.4371(9)
Ba(3)-S(2)	3.3798(4)	In(1)/Zn(5)-Se(5)	2.4371(9)
Ba(3)-S(3)	3.4274(9)	In(1)/Zn(5)-Se(7)	2.4802(16)
Ba(3)-S(3)	3.4274(9)	In(1)/Zn(5)-Se(2)	2.5365(15)
Ba(4)-S(6)	3.0578(12)	In(2)/Zn(6)-Se(4)	2.5211(8)
Ba(4)-S(6)	3.0578(12)	In(2)/Zn(6)-Se(4)	2.5212(8)
Ba(4)-S(6)	3.0578(12)	In(2)/Zn(6)-Se(2)	2.5532(14)
Ba(4)-S(6)	3.0578(12)	In(2)/Zn(6)-Se(1)	2.6445(10)
Ba(4)-S(7)	3.470(3)	In(3)/Zn(7)-Se(5)	2.4794(10)
Ba(4)-S(7)	3.470(3)	In(3)/Zn(7)-Se(3)	2.4942(10)
In(1)-S(5)	2.4757(9)	In(3)/Zn(7)-Se(4)	2.5298(9)
In(1)-S(5)	2.4758(9)	In(3)/Zn(7)-Se(7)	2.5688(9)
In(1)-S(5)	2.4758(9)	In(4)-Se(6)	2.525(2)
In(1)-S(5)	2.4758(9)	In(4)-Se(6)	2.525(2)
In(2)-S(1)	2.4230(9)	In(4)-Se(1)	2.701(6)
In(2)-S(1)	2.4231(9)	In(4)-Se(2)	3.1093(16)
In(2)-S(6)	2.4281(12)	In(4)-Se(2)	3.1093(16)
In(2)-S(4)	2.4830(11)	In(44)-Se(1)	2.729(5)
In(3)-S(3)	2.4366(8)	In(44)-Se(1)	2.729(5)
In(3)-S(3)	2.4366(8)	In(44)-Se(2)	2.800(8)
In(3)-S(2)	2.4812(13)	In(44)-Se(2)	2.799(8)
In(3)-S(7)	2.5887(7)	In(44)-Se(6)	2.5417(18)
In(4)-S(2)	2.5453(11)	In(44)-Se(6)	2.5417(18)

In(4)-S(2)	2.5453(11)	In(44)-Se(6)	2.5417(18)
In(4)-S(2)	2.5453(11)	In(44)-S(2)	2.5620(14)
In(4)-S(2)	2.5453(11)	In(44)-S(2)	2.5642(14)
In(44)-S(7)	2.740(8)		
Zn(1)/In(5)-S(5)	2.3024(10)		
Zn(1)/In(5)-S(1)	2.3168(9)		
Zn(1)/In(5)-S(3)	2.3578(9)		
Zn(1)/In(5)-S(4)	2.4741(6)		