Supporting Information

Table S1 Bond lengths (Å) and angles (deg.).

Dy_3GaS_6[a]		Y_3GaS_6[b]	
Dy(1)-S(1)	2.671(6)	Y(1)-S(1)	2.653(2)
Dy(1)-S(1)#1	2.715(6)	Y(1)-S(1)#1	2.711(2)
Dy(1)-S(2)#2	2.759(6)	Y(1)-S(2)#2	2.756(2)
Dy(1)-S(4)#3	2.790(6)	Y(1)-S(3)	2.784(2)
Dy(1)-S(2)	2.878(6)	Y(1)-S(2)	2.865(2)
Dy(1)-S(3)	2.944(6)	Y(1)-S(4)	2.932(2)
Dy(1)-S(3)#4	2.952(6)	Y(1)-S(4)#3	2.946(2)
Dy(1)-Dy(1)#5	3.941(2)	Y(1)-Y(1)#4	2.932(2)
Dy(2)-S(1)#1	2.723(6)	Y(2)-S(1)#1	2.710(2)
Dy(2)-S(1)#6	2.723(6)	Y(2)-S(1)#5	2.710(2)
Dy(2)-S(1)	2.769(5)	Y(2)-S(1)#6	2.772(2)
Dy(2)-S(1)#7	2.769(5)	Y(2)-S(1)	2.772(2)
Dy(2)-S(2)#8	2.810(6)	Y(2)-S(2)#7	2.795(2)
Dy(2)-S(2)#9	2.810(6)	Y(2)-S(2)#8	2.795(2)
Dy(2)-S(4)#10	3.001(8)	Y(2)-S(3)#8	3.000(3)
Ga(1)-S(4)	2.266(8)	Ga(1)-S(3)#9	2.254(3)
Ga(1)-S(2)#5	2.291(6)	Ga(1)-S(2)#4	2.280(2)
Ga(1)-S(2)	2.291(6)	Ga(1)-S(2)	2.280(2)
Ga(1)-S(3)	2.329(8)	Ga(1)-S(4)	2.320(3)
S(1)-Dy(1)#11	2.715(6)	S(1)-Y(2)#10	2.710(2)
S(1)-Dy(2)#12	2.723(6)	S(1)-Y(1)#11	2.711(2)
S(2)-Dy(1)#13	2.759(6)	S(2)-Y(1)#12	2.756(2)
S(2)-Dy(2)#14	2.810(6)	S(2)-Y(2)#13	2.795(2)
S(3)-Dy(1)#5	2.944(6)	S(3)-Ga(1)#14	2.254(3)
S(3)-Dy(1)#11	2.952(6)	S(3)-Y(1)#4	2.784(2)
S(3)-Dy(1)#15	2.952(6)	S(3)-Y(2)#13	3.000(3)
S(4)-Dy(1)#16	2.790(6)	S(4)-Y(1)#4	2.932(2)
S(4)-Dy(1)#17	2.790(6)	S(4)-Y(1)#15	2.946(2)
S(4)-Dy(2)#18	3.001(8)	S(4)-Y(1)#11	2.946(2)
S(1)-Dy(1)-S(1)#1	76.00(12)	S(1)-Y(1)-S(1)#1	76.26(5)
S(1)-Dy(1)-S(2)#2	103.17(16)	S(1)-Y(1)-S(2)#2	103.15(6)
S(1)#1-Dy(1)-S(2)#2	78.88(16)	S(1)#1-Y(1)-S(2)#2	79.07(8)
S(1)-Dy(1)-S(4)#3	159.2(2)	S(1)-Y(1)-S(3)	159.20(8)
S(1)#1-Dy(1)-S(4)#3	124.6(2)	S(1)#1-Y(1)-S(3)	124.33(9)
S(2)#2-Dy(1)-S(4)#3	80.26(16)	S(2)#2-Y(1)-S(3)	80.24(6)
S(1)-Dy(1)-S(2)	81.98(17)	S(1)-Y(1)-S(2)	81.92(7)
S(1)#1-Dy(1)-S(2)	146.87(16)	S(1)#1-Y(1)-S(2)	47.30(5)

S(2)#2-Dy(1)-S(2)	82.46(9)	S(2)#2-Y(1)-S(2)	82.59(4)
S(4)#3-Dy(1)-S(2)	78.14(18)	S(3)-Y(1)-S(2)	78.15(7)
S(1)-Dy(1)-S(3)	79.93(17)	S(1)-Y(1)-S(4)	79.97(6)
S(1)#1-Dy(1)-S(3)	124.96(19)	S(1)#1-Y(1)-S(4)	124.73(7)
S(2)#2-Dy(1)-S(3)	155.44(19)	S(2)#2-Y(1)-S(4)	155.54(7)
S(4)#3-Dy(1)-S(3)	88.70(16)	S(3)-Y(1)-S(4)	88.73(6)
S(2)-Dy(1)-S(3)	73.79(18)	S(2)-Y(1)-S(4)	73.77(7)
S(1)-Dy(1)-S(3)#4	118.35(19)	S(1)-Y(1)-S(4)#3	118.59(7)
S(1)#1-Dy(1)-S(3)#4	79.09(16)	S(1)#1-Y(1)-S(4)#3	78.79(6)
S(2)#2-Dy(1)-S(3)#4	125.8(2)	S(2)#2-Y(1)-S(4)#3	125.59(8)
S(4)#3-Dy(1)-S(3)#4	72.9(2)	S(3)-Y(1)-S(4)#3	72.76(8)
S(2)-Dy(1)-S(3)#4	133.72(15)	S(2)-Y(1)-S(4)#3	133.60(6)
S(3)-Dy(1)-S(3)#4	70.27(8)	S(4)-Y(1)-S(4)#3	70.27(4)
S(1)-Dy(1)-Dy(1)#5	127.78(12)	S(1)-Y(1)-Y(1)#4	127.75(4)
S(1)#1-Dy(1)-Dy(1)#5	127.06(11)	S(1)#1-Y(1)-Y(1)#4	126.80(4)
S(2)#2-Dy(1)-Dy(1)#5	125.33(11)	S(2)#2-Y(1)-Y(1)#4	125.32(4)
S(4)#3-Dy(1)-Dy(1)#5	45.07(12)	S(3)-Y(1)-Y(1)#4	45.08(4)
S(2)-Dy(1)-Dy(1)#5	86.03(11)	S(2)-Y(1)-Y(1)#4	85.86(4)
S(3)-Dy(1)-Dy(1)#5	47.98(10)	S(4)-Y(1)-Y(1)#4	47.89(4)
S(3)#4-Dy(1)-Dy(1)#5	48.12(10)	S(4)#3-Y(1)-Y(1)#4	48.14(4)
S(1)#1-Dy(2)-S(1)#6	78.9(2)	S(1)#1-Y(2)-S(1)#5	79.25(9)
S(1)#1-Dy(2)-S(1)	74.28(12)	S(1)#1-Y(2)-S(1)#6	121.69(5)
S(1)#6-Dy(2)-S(1)	121.55(10)	S(1)#5-Y(2)-S(1)#6	74.32(5)
S(1)#1-Dy(2)-S(1)#7	121.55(10)	S(1)#1-Y(2)-S(1)	74.32(5)
S(1)#6-Dy(2)-S(1)#7	74.28(11)	S(1)#5-Y(2)-S(1)	121.69(5)
S(1)-Dy(2)-S(1)#7	77.3(2)	S(1)#6-Y(2)-S(1)	77.17(8)
S(1)#1-Dy(2)-S(2)#8	158.07(18)	S(1)#1-Y(2)-S(2)#7	97.05(6)
S(1)#6-Dy(2)-S(2)#8	97.27(16)	S(1)#5-Y(2)-S(2)#7	157.74(8)
S(1)-Dy(2)-S(2)#8	124.4(2)	S(1)#6-Y(2)-S(2)#7	124.51(9)
S(1)#7-Dy(2)-S(2)#8	77.11(17)	S(1)-Y(2)-S(2)#7	77.38(7)
S(1)#1-Dy(2)-S(2)#9	97.27(16)	S(1)#1-Y(2)-S(2)#8	157.74(8)
S(1)#6-Dy(2)-S(2)#9	158.07(18)	S(1)#5-Y(2)-S(2)#8	97.05(6)
S(1)-Dy(2)-S(2)#9	77.11(16)	S(1)#6-Y(2)-S(2)#8	77.38(7)
S(1)#7-Dy(2)-S(2)#9	124.4(2)	S(1)-Y(2)-S(2)#8	124.51(9)
S(2)#8-Dy(2)-S(2)#9	78.1(2)	S(2)#7-Y(2)-S(2)#8	78.01(8)
S(1)#1-Dy(2)-S(4)#10	82.27(17)	S(1)#1-Y(2)-S(3)#8	81.99(7)
S(1)#6-Dy(2)-S(4)#10	82.27(18)	S(1)#5-Y(2)-S(3)#8	81.99(7)
S(1)-Dy(2)-S(4)#10	141.29(11)	S(1)#6-Y(2)-S(3)#8	141.38(4)
S(1)#7-Dy(2)-S(4)#10	141.29(11)	S(1)-Y(2)-S(3)#8	141.38(4)
S(2)#8-Dy(2)-S(4)#10	75.81(15)	S(2)#7-Y(2)-S(3)#8	75.75(6)
S(2)#9-Dy(2)-S(4)#10	75.81(15)	S(2)#8-Y(2)-S(3)#8	75.75(6)
S(4)-Ga(1)-S(2)#5	117.7(2)	S(3)#9-Ga(1)-S(2)#4	117.84(7)
S(4)-Ga(1)-S(2)	117.7(2)	S(3)#9-Ga(1)-S(2)	117.84(7)
S(2)#5-Ga(1)-S(2)	101.3(3)	S(2)#4-Ga(1)-S(2)	101.02(11)

S(4)-Ga(1)-S(3)	119.8(3)	S(3)#9-Ga(1)-S(4)	119.61(11)
S(2)#5-Ga(1)-S(3)	98.3(2)	S(2)#4-Ga(1)-S(4)	98.31(8)
S(2)-Ga(1)-S(3)	98.3(2)	S(2)-Ga(1)-S(4)	98.31(8)
Dy(1)-S(1)-Dy(1)#11	101.41(17)	Y(1)-S(1)-Y(2)#10	140.26(8)
Dy(1)-S(1)-Dy(2)#12	139.7(2)	Y(1)-S(1)-Y(1)#11	101.53(7)
Dy(1)#11-S(1)-Dy(2)#12	104.9(2)	Y(2)#10-S(1)-Y(1)#11	104.74(8)
Dy(1)-S(1)-Dy(2)	104.8(2)	Y(1)-S(1)-Y(2)	104.63(8)
Dy(1)#11-S(1)-Dy(2)	103.03(17)	Y(2)#10-S(1)-Y(2)	98.15(6)
Dy(2)#12-S(1)-Dy(2)	98.36(17)	Y(1)#11-S(1)-Y(2)	102.57(7)
Ga(1)-S(2)-Dy(1)#13	115.9(2)	Ga(1)-S(2)-Y(1)#12	115.71(9)
Ga(1)-S(2)-Dy(2)#14	89.02(18)	Ga(1)-S(2)-Y(2)#13	89.18(7)
Dy(1)#13-S(2)-Dy(2)#14	100.86(17)	Y(1)#12-S(2)-Y(2)#13	100.83(7)
Ga(1)-S(2)-Dy(1)	94.55(19)	Ga(1)-S(2)-Y(1)	94.66(7)
Dy(1)#13-S(2)-Dy(1)	140.7(2)	Y(1)#12-S(2)-Y(1)	140.53(7)
Dy(2)#14-S(2)-Dy(1)	104.1(2)	Y(2)#13-S(2)-Y(1)	104.37(7)
Ga(1)-S(3)-Dy(1)#5	92.1(2)	Ga(1)#14-S(3)-Y(1)#4	112.25(8)
Ga(1)-S(3)-Dy(1)	92.1(2)	Ga(1)#14-S(3)-Y(1)	112.25(8)
Dy(1)#5-S(3)-Dy(1)	84.0(2)	Y(1)#4-S(3)-Y(1)	89.83(9)
Ga(1)-S(3)-Dy(1)#11	114.2(2)	Ga(1)#14-S(3)-Y(2)#13	131.76(11)
Dy(1)#5-S(3)-Dy(1)#11	153.3(3)	Y(1)#4-S(3)-Y(2)#13	101.20(8)
Dy(1)-S(3)-Dy(1)#11	89.98(7)	Y(1)-S(3)-Y(2)#13	101.20(8)
Ga(1)-S(3)-Dy(1)#15	114.2(2)	Ga(1)-S(4)-Y(1)#4	92.04(8)
Dy(1)#5-S(3)-Dy(1)#15	89.98(7)	Ga(1)-S(4)-Y(1)	92.04(8)
Dy(1)-S(3)-Dy(1)#15	153.3(3)	Y(1)#4-S(4)-Y(1)	84.21(8)
Dy(1)#11-S(3)-Dy(1)#15	83.8(2)	Ga(1)-S(4)-Y(1)#15	114.16(8)
Ga(1)-S(4)-Dy(1)#16	111.9(2)	Y(1)#4-S(4)-Y(1)#15	89.95(4)
Ga(1)-S(4)-Dy(1)#17	111.9(2)	Y(1)-S(4)-Y(1)#15	153.37(10)
Dy(1)#16-S(4)-Dy(1)#17	89.9(2)	Ga(1)-S(4)-Y(1)#11	114.16(8)
Ga(1)-S(4)-Dy(2)#18	131.8(4)	Y(1)#4-S(4)-Y(1)#11	153.37(10)
Dy(1)#16-S(4)-Dy(2)#18	101.5(2)	Y(1)-S(4)-Y(1)#11	89.95(4)
Dy(1)#17-S(4)-Dy(2)#18	101.5(2)	Y(1)#15-S(4)-Y(1)#11	83.72(8)

[a]Symmetry codes: (#1) x,-y+1,z-1/2 ; (#2) -x+1/2,-y+1/2,z-1/2 ; (#3) x,y,z-1 ; (#4) -x,-y+1,z-1/2 ; (#5) -x,y,z ; (#6) -x+1,-y+1,z-1/2 ; (#7) -x+1,y,z ; (#8) x+1/2,y+1/2,z ; (#9) -x+1/2,y+1/2,z ; (#10) x+1/2,y+1/2,z-1 ; (#11) x,-y+1,z+1/2 ; (#12) -x+1,-y+1,z+1/2 ; (#13) -x+1/2,-y+1/2,z+1/2 ; (#14) x-1/2,y-1/2,z ; (#15) -x,-y+1,z+1/2 ; (#16) x,y,z+1 ; (#17) -x,y,z+1 ; (#18) x-1/2,y-1/2,z+1.

[b]Symmetry codes: (#1) x,-y,z-1/2 ; (#2) -x+1/2,-y+1/2,z-1/2 ; (#3) -x,-y,z-1/2 ; (#4) -x,y,z ; (#5) -x+1,-y,z-1/2 ; (#6) -x+1,y,z ; (#7) -x+1/2,y-1/2,z ; (#8) x+1/2,y-1/2,z ; (#9) x,y,z+1 ; (#10) -x+1,-y,z+1/2 ; (#11) x,-y,z+1/2 ; (#12) -x+1/2,-y+1/2,z+1/2 ; (#13) x-1/2,y+1/2,z ; (#14) x,y,z-1 ; (#15) -x,-y,z+1/2.

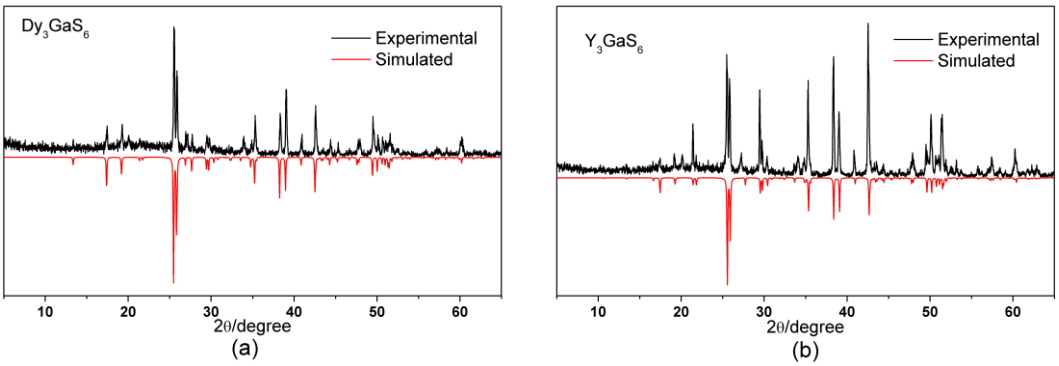

Fig. S1 The experimental and simulated X-ray powder diffraction patterns for Dy_3GaS_6 (a) and Y_3GaS_6 (b).

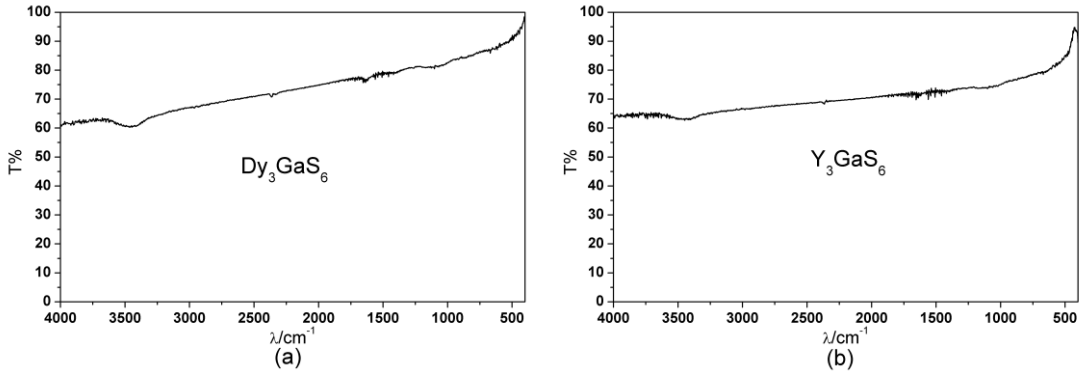

Fig. S2 The IR spectra of Dy_3GaS_6 (a) and Y_3GaS_6 (b).

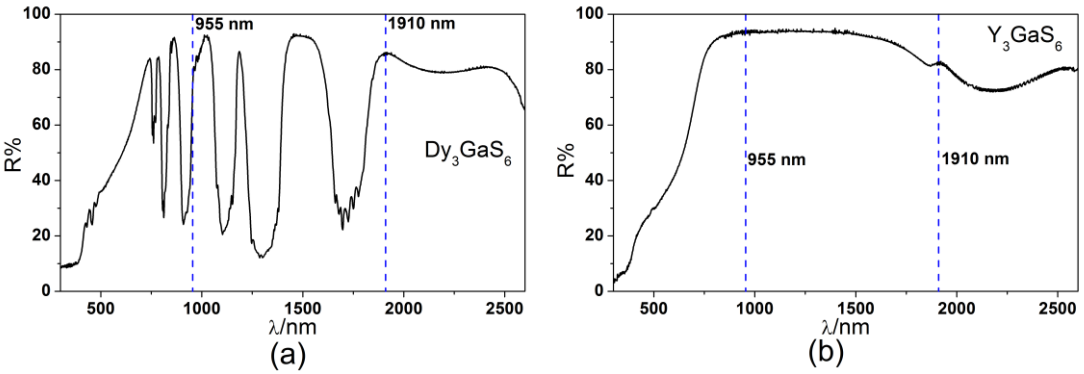

Fig. S3 The UV-Vis-NIR diffuse reflectance spectra of Dy_3GaS_6 (a) and Y_3GaS_6 (b).

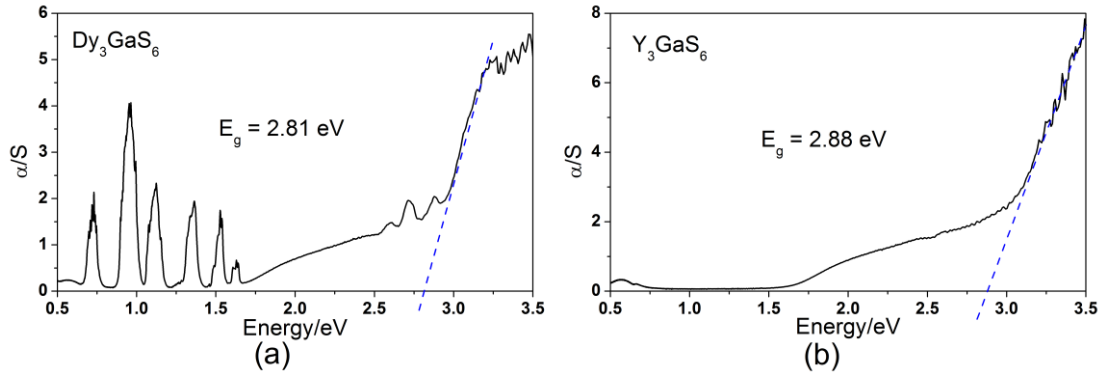

Fig. S4 The UV-Vis-NIR absorption spectra of Dy_3GaS_6 (a) and Y_3GaS_6 (b).

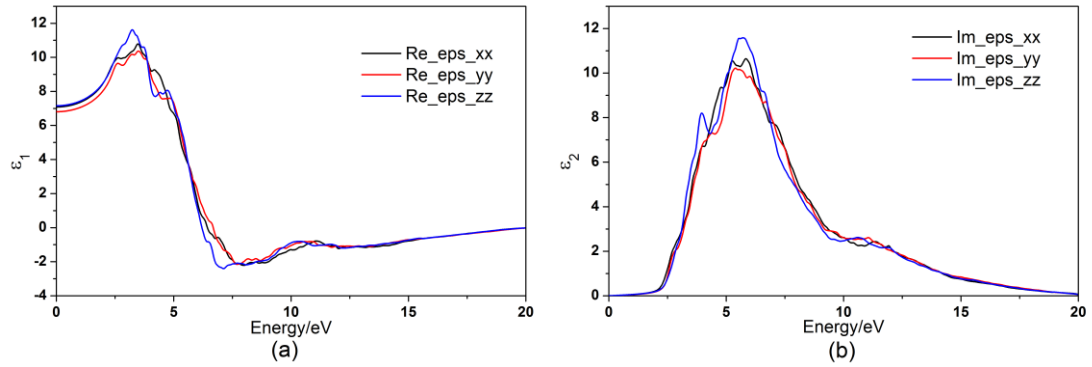

Fig. S5 Energy dependences of the real part ε_1 (a) and the imaginary part ε_2 (b) of Dy_3GaS_6.

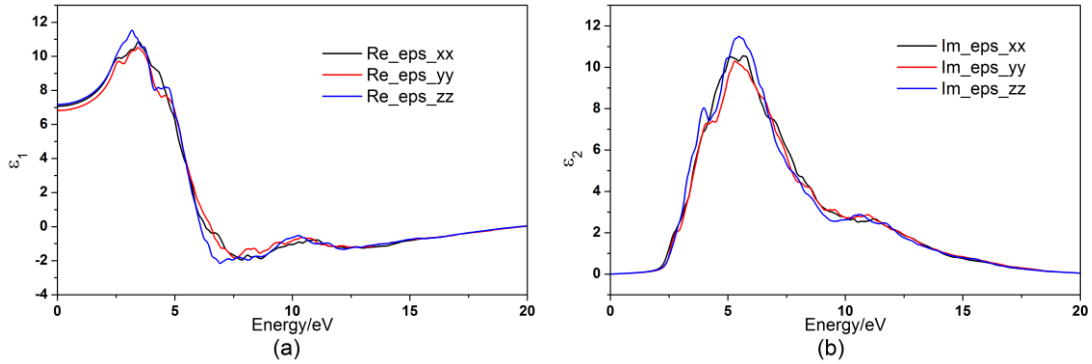

Fig. S6 Energy dependences of the real part ε_1 (a) and the imaginary part ε_2 (b) of Y_3GaS_6.

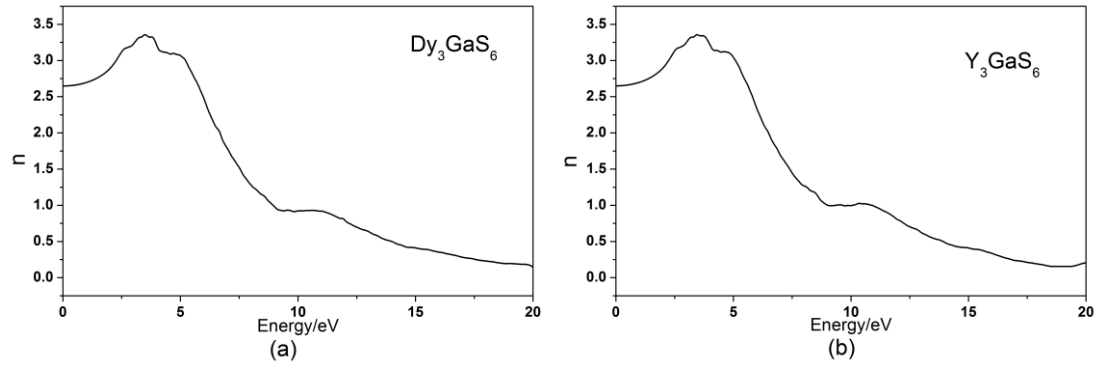

Fig. S7 Variations of the calculated average refractive index n of Dy_3GaS_6 (a) and Y_3GaS_6 (b).

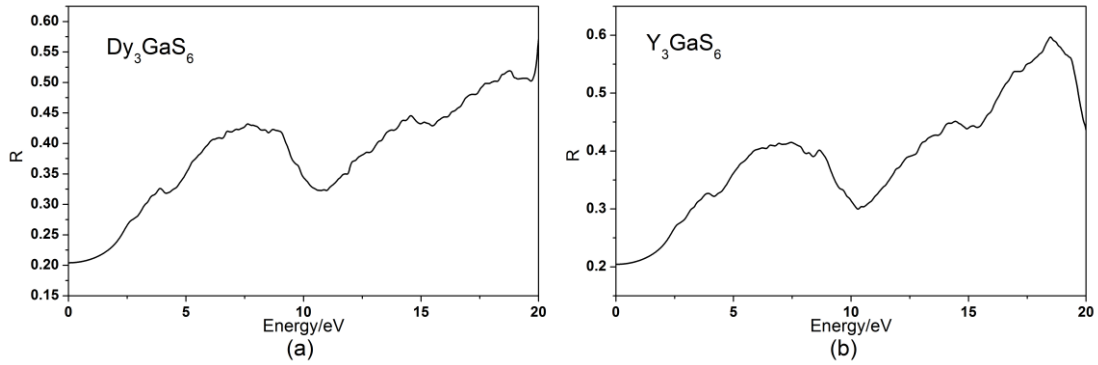

Fig. S8 Calculated average reflectivity R of Dy_3GaS_6 (a) and Y_3GaS_6 (b).

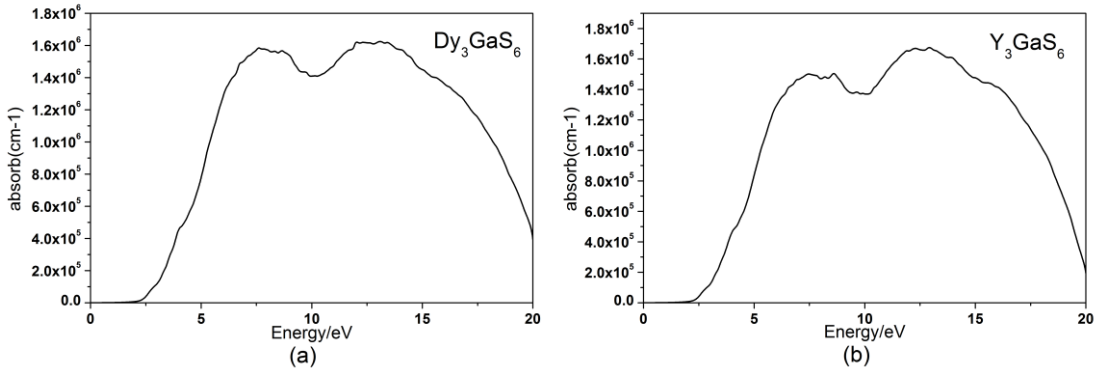

Fig. S9 Calculated average absorption coefficient *I* of Dy_3GaS_6 (a) and Y_3GaS_6 (b).

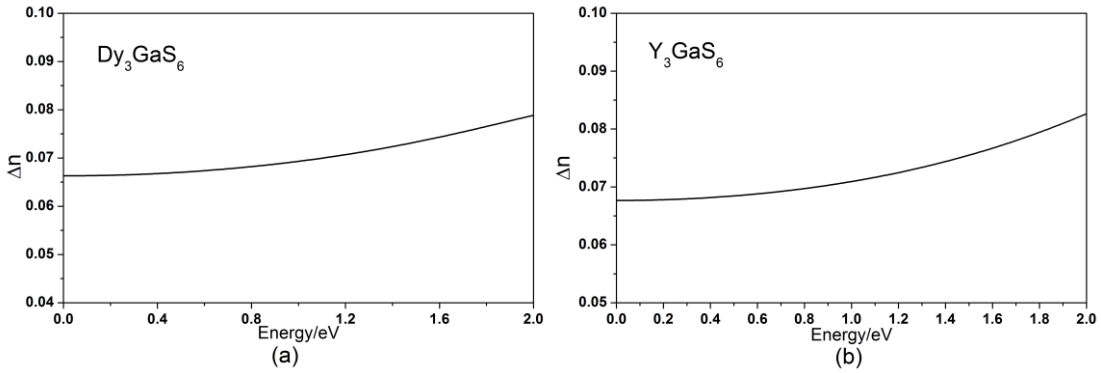

Fig. S10 Calculated birefringence Δn of Dy_3GaS_6 (a) and Y_3GaS_6 (b).