Supporting Information

$Ga(IO_3)_3$: A mid-IR nonlinear optical iodate with balanced performance between band gap and second harmonic generation response

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1. Supplementary Tables.

Table S1. Crystal data and structure refinements of $Ga(IO_3)_3$.

Formula	$Ga(IO_3)_3$
Temperature (K)	290
Crystal system	Hexagonal
Space group	$P6_3$
a (Å)	9.0924 (5)
b (Å)	9.0924 (5)
c (Å)	5.2862 (8)
α (°)	90
β (°)	90
γ (°)	120
Z	2
V (Å^3)	378.47 (6)
μ (mm^{-1})	15.90
$R[F^2 > 2\sigma (F^2)]^a$	0.016
$wR(F^2)$	0.037

^{a} $R_1 = \Sigma||F_o| - |F_c||/\Sigma|F_o|$, $\omega R_2 = [\Sigma\omega(F_o^2 - F_c^2)_2/\Sigma\omega(F_o^2)^2]^{1/2}$

Table S2. Selected bond lengths (Å) of $Ga(IO_3)_3$.

Bond	Length/Å
I1-O1	1.847(5)
I1-O2	1.804(6)
I1-O3	1.806(6)
Ga1-O1	1.980(2)
Ga1-O1	1.980(2)
Ga1-O1	1.980(2)
Ga1-O3	1.960(2)
Ga1-O3	1.960(2)
Ga1- O3	1.961(2)

Table S3. Selected bond angles (°) of $Ga(IO_3)_3$.

Angle	(°)
O1-I1-O2	99.61(12)
O1-I1-O3	94.68(11)
O2-I1-O3	96.61(12)
O1-Ga1-O1	90.75(11)
O1-Ga1-O3	169.98(10)
O1-Ga1-O3	85.24(10)
O1-Ga1-O3	98.46(10)
O1-Ga1-O3	98.45(10)
O3-Ga1-O3	86.28(10)

Table S4. Space group, SHG responses, band gap, birefringence for the iodate materials only including IO_3^- unit.

Compounds	Space group	SHG effect (× KDP)	Band gap (eV)	Birefringence	Ref
$LiMg(IO_3)_3$	$P6_3$	24	4.34	0.22 @ 1064 nm[a]	S1
$K_2Mg(IO_3)_4(H_2O)_2$	$I2$	1.4	4.37	0.021 @ 1064 nm[a]	S2
$Ba_2[MoO_3(OH)(IO_3)_2]IO_3$	$P2_1$	8	3.78	0.225 @ 1064 nm[a]	S3
$CdIO_3F$	$P2_12_12_1$	6.2	4.22	0.072 @ 1064 nm[a] 0.068 @ 546.1 nm[b]	S4
$K_5(W_3O_9F_4)(IO_3)$	Pm	11	3.83	0.083 @ 1064 nm[a]	S5
$(H_3O)HCs_2Nb(IO_3)_9$	$P2_1$	6	3.58	0.052 @ 1064 nm[a]	S6
$LiZn(IO_3)_3$	$P6_3$	14	4.21	0.27 @ 1064 nm[a]	S7
$LiCd(IO_3)_3$	$P6_3$	12	4.18	0.27 @ 1064 nm[a]	S7
$NaVO_2(IO_3)_2(H_2O)$	$P2_1$	20	3.06	0.21 @ 1064 nm[a]	S8
$K_2Zn(IO_3)_4(H_2O)_2$	$I2$	2.3	4.35	0.018 @ 1064 nm[a]	S2
$BaNbO(IO_3)_5$	Cc	14	3.64	0.035 @ 1064 nm[a]	S9
$NH_4[MoO_3(IO_3)]$	$Pna2_1$	4.7	3.26	0.083 @ 1064 nm[a]	S10
$KRb[(MoO_3)_2(IO_3)_2]$	Cc	8.5	3.32	0.146 @ 1064 nm[a]	S10
$Ce(IO_3)_4$	$R3c$	0.9	2.17	0.049 @ 546 nm[b]	S11
$Y(IO_3)_2F$	$P6_5$	2	3.91	0.041 @ 1064 nm[a]	S12
β-$Sc(IO_3)_3$	$P6_3$	16	4.52	0.253 @ 546 nm[a] 0.219 @ 546 nm[b]	S13
$Ce(IO_3)_2F_2\cdot H_2O$	$Ima2$	3	2.6	0.046 @ 1064 nm[a]	S14
$Sn(IO_3)_2F_2$	$P2_1$	3	4.08	0.234 @ 1064 nm[a]	S15
$Bi(IO_3)F_2$	$C2$	11.5	3.97	0.209 @ 1064 nm[a]	S16
$Bi_2Te(IO_3)O_5Cl$	Cc	3	3.6	0.091 @ 1064 nm[a]	S17
$[GaF(H_2O)][IO_3F]$	$Pca2_1$	10	4.34	0.142 @ 1064 nm[a]	S18
α-$Ba_2[GaF_4(IO_3)_2](IO_3)$	$Pna2_1$	~ 6	4.61	0.126 @ 1064 nm[a]	S19
β-$Ba_2[GaF_4(IO_3)_2](IO_3)$	$P2_1$	~ 6	4.35	0.135 @ 1064 nm[a]	S19
$Ga(IO_3)_3$	$P6_3$	13	3.94	0.187 @ 1064 nm[a] 0.159[b]	This work

[a.] calculated birefringence; [b.] experimented birefringence.

Table S5. Properties of NLO gallium iodates.

Compounds	Space group	SHG effect (× KDP)	Band gap (eV)	Birefringence	Ref
$[GaF(H_2O)][IO_3F]$	$Pca2_1$	10	4.34	0.142 @ 1064 nm[a]	S18
α-$Ba_2[GaF_4(IO_3)_2](IO_3)$	$Pna2_1$	~ 6	4.61	0.126 @ 1064 nm[a]	S19
β-$Ba_2[GaF_4(IO_3)_2](IO_3)$	$P2_1$	~ 6	4.35	0.135 @ 1064 nm[a]	S19
$Ga(IO_3)_3$	$P6_3$	13	3.94	0.187 @ 1064 nm[a] 0.159[b]	This work

[a.] calculated data; [b.] experimented data.

Table S6. Calculation of the dipole moment for GaO_6 and IO_3 polyhedrons and the net dipole moment for a unit cell, and BSI and GII indices of $Ga(IO_3)_3$.

$Ga(IO_3)_3$ (Z = 2)					
Species	Valence of central atom	Dipole moment (D = Debye)			
		x(a)	y(b)	z(c)	total magnitude
Ga1O_6	3.1377	0	0	-1.5237	1.5237
Ga1O_6	3.1377	0	0	-1.5237	1.5237
I1O_3	4.9390	6.3867	-15.6911	-37.2953	40.9627
I1O_3	4.9390	5.0027	7.9833	-32.4618	33.8013
I1O_3	4.9390	-23.0483	2.0361	-27.6529	36.0563
I1O_3	4.9390	-6.3867	15.6911	-37.2953	40.9627
I1O_3	4.9390	5.0027	-7.9833	-32.4618	33.8013
I1O_3	4.9390	23.0483	-2.0361	-27.6529	36.0563
Net dipole moment (GaO_6)		0	0	-3.0474	
Net dipole moment (IO_3)		0	0	-194.82	
Net dipole moment (a unit cell)		0	0	-197.8674	
Cell volume	1163.45 Å^3				
Dipole moment density (IO_3)	194.82/1163.45=0.167 D/Å^3				
Dipole moment density (a unit cell)	197.8674/1163.45= 0.170 D/Å^3				
Bond Strain Index (BSI)	0.190 vu				
Global Instability Index (GII)	0.178 vu				

2. Supplementary Figures.

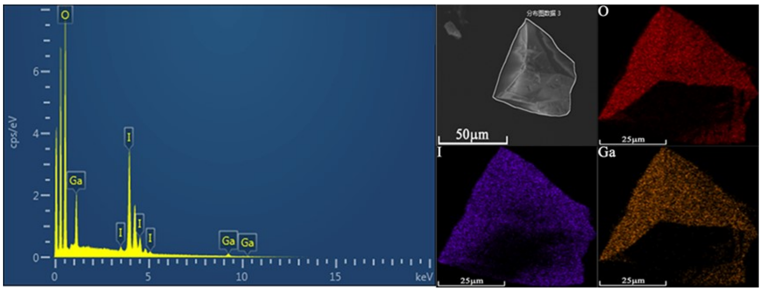

Figure S1. The EDS spectrum of $Ga(IO_3)_3$.

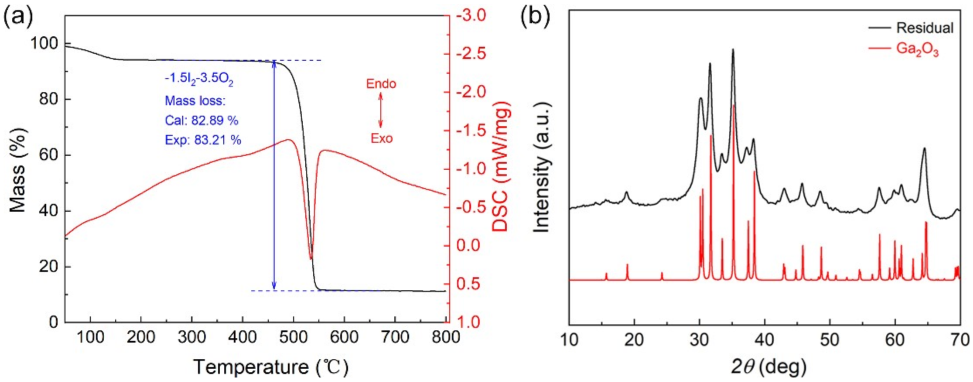

Figure S2. (a) The TG and DSC curves and (b) PXRD pattern of the residual of $Ga(IO_3)_3$.

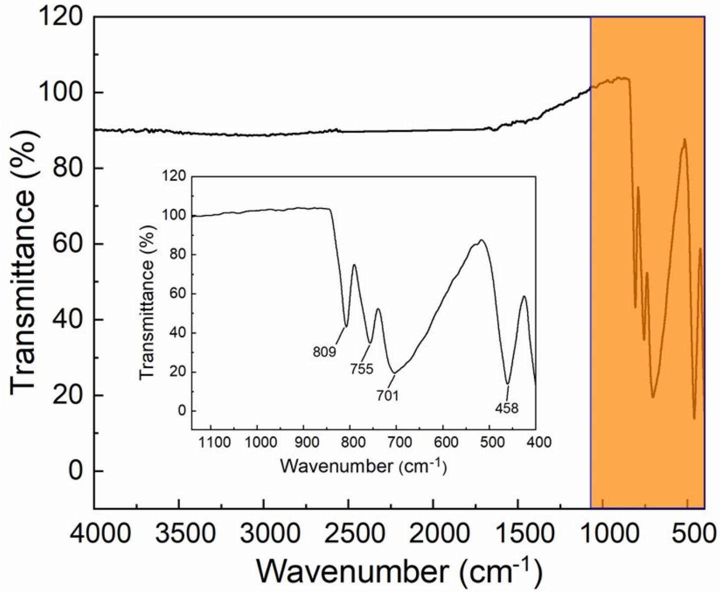

Figure S3. The IR spectrum of $Ga(IO_3)_3$.

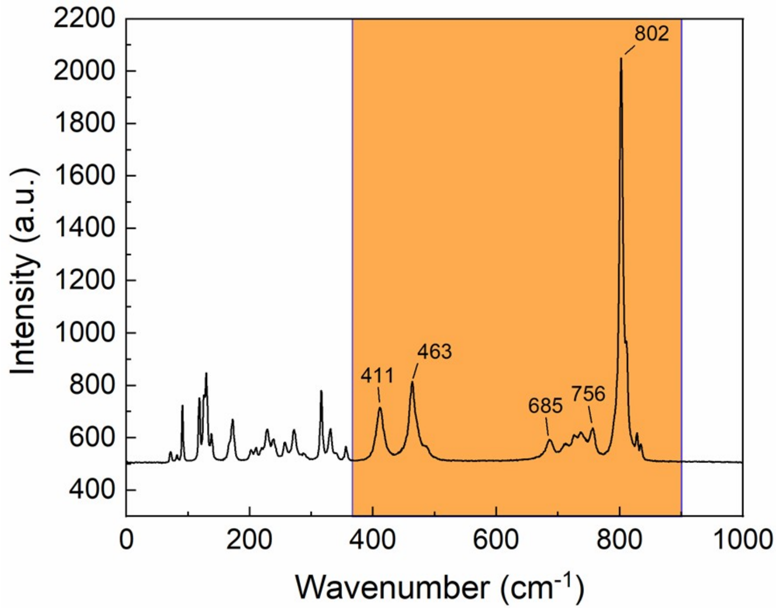

Figure S4. The Raman spectrum of $Ga(IO_3)_3$.

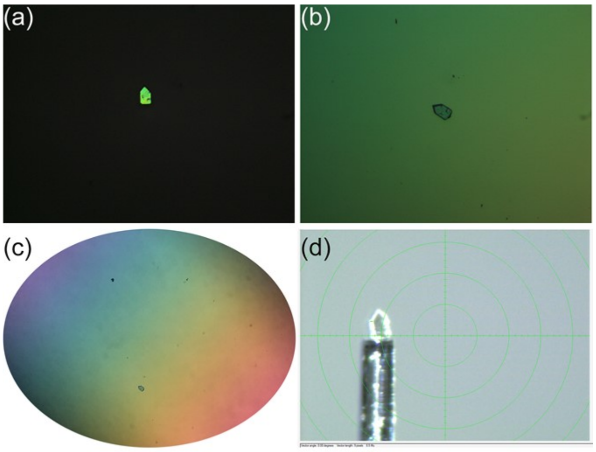

Figure S5. Birefringence measurement of $Ga(IO_3)_3$; (a) the original crystal; (b) the crystal in the extinction state; (c) the crystal interference color observed under the microscope and (d) the photographs of crystal thickness.

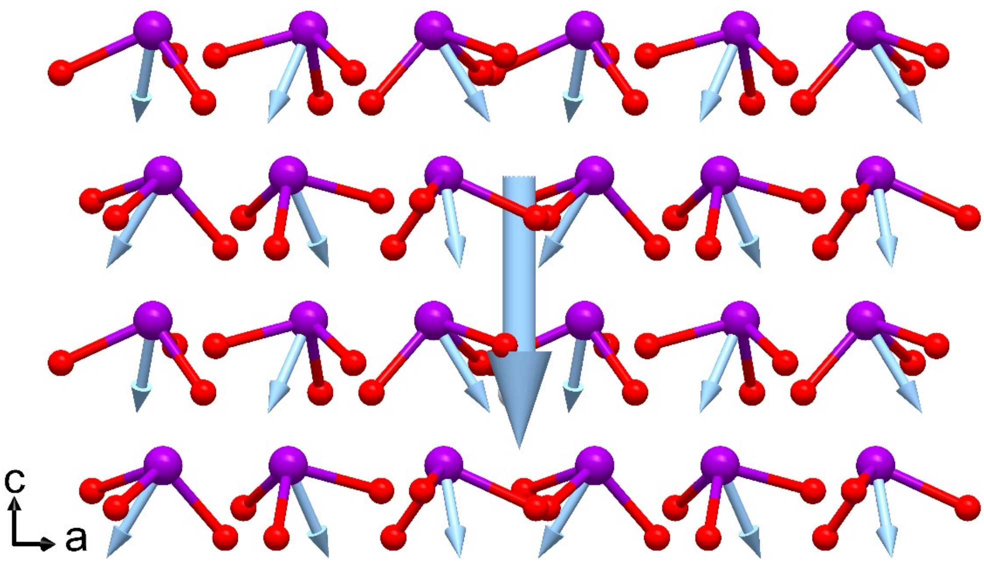

Figure S6. The direction of dipole moments of IO_3^- units in $Ga(IO_3)_3$.

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