Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

An Effective Pathway to Design and Synthesize UV Birefringent Crystals via Rational Assemble of π-conjugated $[CO_3]^{2-}$ and $[NO_3]^-$ Triangles

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Table of Contents

Table S1. Crystal data and structure refinement for $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S3**

Table S2. Atomic coordinates ($\times10^4$), equivalent isotropic displacement parameters ($Å^2 \times 10^3$) and bond valence sums (BVS) for $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. **S4**

Table S3. Atomic coordinates ($\times10^4$), equivalent isotropic displacement parameters ($Å^2 \times 10^3$) and bond valence sums (BVS) for $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S4**

Table S4. Bond lengths [Å] and angles [°] for $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. **S5**

Table S5. Bond lengths [Å] and angles [°] for $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S6**

Table S6. Hydrogen coordinates ($\times 10^4$) and isotropic displacement parameters ($Å^2 \times 10^3$) for $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. **S7**

Table S7. Hydrogen coordinates ($\times 10^4$) and isotropic displacement parameters ($Å^2 \times 10^3$) for $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S7**

Table S8. The refractive indices and birefringence values of $Na_3Rb_6(CO_3)_3(NO_3)_2X{\cdot}6H_2O$ (X=Br and Cl). **S7**

Table S9. Birefringences and band gaps of selected carbonates and nitrates. **S8**

Figure S1. The photos of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$ crystals. **S9**

Figure S2. Experimental and theoretical PXRD patterns of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S9**

Figure S3. The unit cell of $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. **S9**

Figure S4. The asymmetric unit of $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. **S10**

Figure S5. Coordination environments of cations in $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. **S10**

Figure S6. A $[BrRb_6]^{5+}$ polyhedron in $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. **S10**

Figure S7. TG-DSC curves of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S11**

Figure S8. PXRD patterns of residues after decomposition for (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S11**

Figure S9. IR spectra of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S11**

Figure S10. The calculated band gaps and PDOS plots for (a, b) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (c, d) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. **S12**

Figure S11. The selected crystal before and after compensatory rotation for birefringence test. **S12**

References **S13**

Table S1. Crystal data and structure refinement for $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$

Formula	**$Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$**	**$Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$**
Formula weight	1073.85	1029.39
Crystal system	Hexagonal	
Space group	*P*6$_3$/*mcm*	
Temperature/K	293(2) K	
a(Å)	9.6086(4)	9.5886(2)
b(Å)	9.6086(4)	9.5886(2)
c(Å)	15.8641(15)	15.8392(4)
V/Å^3	1268.43(14)	1261.17(6)
Z	2	2
ρ(calcd)Mg/m^3	2.812	2.711
μ/mm^{-1}	13.199	11.797
F(000)	1004	968
λ (Å)	0.71073	0.71073
Completeness to theta = 25.242°	99.7 %	100.0 %
No. of reflections	603	560
No. of parameters	40	40
$\Delta\rho_{max}$	0.553	0.299
$\Delta\rho_{min}$	-0.562	-0.258
R_1/wR_2 (I > 2σ(I))[a]	0.0327/ 0.0691	0.0199/0.0357
R_1/wR_1 (all data)[a]	0.0344/ 0.0696	0.0313/ 0.0386

[a]$R_1=\Sigma||F_o|-|F_c||/\Sigma|F_o|$ and $wR_2=[\Sigma w(F_o^2-F_c^2)^2/\Sigma wF_o^4]^{1/2}$ for $F_o^2>2\sigma(F_o^2)$

Table S2. Atomic coordinates (×10⁴), equivalent isotropic displacement parameters (Å² × 10³) and bond valence sums (BVS) for $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$. U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

atom	x	y	z	U(eq) Å²	BVS
Rb(1)	2858(1)	2858(1)	1202(1)	31(1)	+1.130
Na(1)	3776(4)	0	2500	34(1)	+1.178
N(1)	6667	3333	2500	21(2)	+4.817
C(1)	0	0	2500	18(2)	+4.282
C(2)	6667	3333	0	21(2)	+3.992
O(1)	6036(4)	6036(4)	1052(2)	30(1)	−0.463
O(2)	1333(4)	0(4)	2500(2)	33(1)	−2.007
O(3)	5122(5)	2561(2)	0	31(1)	−1.592
O(4)	5252(5)	3076(5)	2500	32(1)	−2.105
Br(1)	0	0	0	31(1)	−1.307

Table S3. Atomic coordinates (×10⁴), equivalent isotropic displacement parameters (Å² × 10³) and bond valence sums (BVS) for $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$. U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

atom	x	y	z	U_{eq} Å²	BVS
Rb(1)	2741(1)	2741(1)	1193(1)	25(1)	+1.089
Na(1)	3703(2)	0	2500	22(1)	+1.214
N(1)	6667	3333	2500	21(1)	+4.960
C(1)	0	0	2500	19(2)	+3.984
C(2)	6667	3333	0	22(1)	+3.974
O(1)	5999(3)	5999(3)	1052(1)	32(1)	−0.439
O(2)	1340(3)	0	2500	25(1)	−2.055
O(3)	5118(3)	2559(1)	0	30(1)	−1.575
O(4)	5264(3)	3093(3)	2500	32(1)	−2.089
Cl(1)	0	0	0	27(1)	−1.233

Table S4. Bond lengths [Å] and angles [°] for $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$.

Rb(1)-O(3)#1	3.012(2)	N(1)-Na(1)#6	3.0127(14)
Rb(1)-O(3)	3.012(2)	C(1)-O(2)#13	1.281(2)
Rb(1)-O(4)	3.015(3)	C(1)-O(2)#3	1.281(2)
Rb(1)-O(4)#2	3.015(3)	C(1)-O(2)	1.281(2)
Rb(1)-O(1)	3.062(4)	C(1)-Rb(1)#12	3.4324(6)
Rb(1)-O(2)#3	3.147(4)	C(1)-Rb(1)#11	3.4324(6)
Rb(1)-O(2)	3.147(4)	C(1)-Rb(1)#3	3.4324(6)
Rb(1)-Br(1)	3.3435(6)	C(1)-Rb(1)#13	3.4324(6)
Rb(1)-O(1)#4	3.413(3)	C(1)-Rb(1)#14	3.4324(6)
Rb(1)-O(1)#5	3.413(3)	C(2)-O(3)#4	1.285(4)
Rb(1)-Na(1)#6	3.834(3)	C(2)-O(3)#15	1.285(4)
Rb(1)-Na(1)#3	3.871(2)	C(2)-O(3)	1.285(4)
Rb(1)-H(1)	3.2294	O(1)-Na(1)#6	2.303(4)
Na(1)-O(1)#7	2.303(4)	O(1)-Rb(1)#16	3.413(3)
Na(1)-O(1)#4	2.303(4)	O(1)-Rb(1)#15	3.413(3)
Na(1)-O(2)	2.347(4)	O(1)-H(1)	0.8918
Na(1)-O(4)#8	2.560(4)	O(2)-Rb(1)#12	3.147(4)
Na(1)-O(4)	2.560(4)	O(2)-Rb(1)#11	3.147(4)
Na(1)-O(4)#4	2.683(5)	O(2)-Rb(1)#13	3.147(4)
Na(1)-O(4)#9	2.683(5)	O(3)-Rb(1)#17	3.012(2)
Na(1)-N(1)#10	3.0127(14)	O(4)-Na(1)#6	2.683(5)
Na(1)-Rb(1)#7	3.834(3)	O(4)-Rb(1)#12	3.015(3)
Na(1)-Rb(1)#4	3.834(3)	Br(1)-Rb(1)#18	3.3435(6)
Na(1)Rb(1)#11	3.871(2)	Br(1)-Rb(1)#1	3.3435(6)
Na(1)Rb(1)#12	3.871(2)	Br(1)-Rb(1)#3	3.3435(6)
N(1)-O(4)#4	1.254(4)	Br(1)-Rb(1)#17	3.3435(6)
N(1)-O(4)#6	1.254(4)	Br(1)-Rb(1)#11	3.3435(6)
N(1)-O(4)	1.254(4)		

Symmetry transformations used to generate equivalent atoms:
#1 x-y,x,-z #2 y,x,z #3 -y,x-y,z
#4 -y+1,x-y,z #5 -x+y,-x+1,z #6 -x+y+1,-x+1,-z+1/2
#7 -y+1,x-y,-z+1/2 #8 x-y,-y,-z+1/2
#9 -x+1,-x+y,z #10 y,x-1,z #11 -x+y,-x,z
#12 x,y,-z+1/2 #13 -x+y,-x,-z+1/2
#14 -y,x-y,-z+1/2 #15 -x+y+1,-x+1,z
#16 -y+1,x-y+1,z #17 y,-x+y,-z #18 -x,-y,-z

Table S5. Bond lengths [Å] and angles [°] for $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$.

Rb(1)-O(3)	3.0322(16)	N(1)-Na(1)#6	3.0344(7)
Rb(1)-O(3)#1	3.0322(15)	C(1)-O(2)#13	1.285(3)
Rb(1)-O(4)#2	3.0717(18)	C(1)-O(2)#3	1.285(3)
Rb(1)-O(4)	3.0718(18)	C(1)-O(2)	1.285(3)
Rb(1)-O(2)	3.0770(3)	C(1)-Rb(1)#12	3.3458(3)
Rb(1)-O(2)#3	3.0770(3)	C(1)-Rb(1)#11	3.3458(3)
Rb(1)-O(1)	3.132(2)	C(1)-Rb(1)#3	3.3458(3)
Rb(1)-Cl(1)	3.2373(3)	C(1)-Rb(1)#13	3.3458(3)
Rb(1)-O(1)#4	3.4050(18)	C(1)-Rb(1)#14	3.3458(3)
Rb(1)-O(1)#5	3.4050(18)	C(2)-O(3)#5	1.286(2)
Rb(1)-Na(1)#3	3.8039(11)	C(2)-O(3)#15	1.286(2)
Rb(1)-Na(1)#6	3.9884(16)	C(2)-O(3)	1.286(2)
Na(1)-O(2)	2.266(4)	O(1)-Na(1)#6	2.312(2)
Na(1)-O(1)#7	2.312(2)	O(1)-Rb(1)#16	3.4050(18)
Na(1)-O(1)#5	2.312(2)	O(1)-Rb(1)#15	3.4050(18)
Na(1)-O(4)	2.569(2)	O(1)-H(1)	0.8664
Na(1)-O(4)#8	2.569(2)	O(2)-Rb(1)#11	3.0770(3)
Na(1)-O(4)#5	2.715(3)	O(2)-Rb(1)#12	3.0770(3)
Na(1)-O(4)#9	2.715(3)	O(2)-Rb(1)#13	3.0770(3)
Na(1)-N(1)#10	3.0345(7)	O(3)-Rb(1)#17	3.0322(15)
Na(1)-Rb(1)#11	3.8039(11)	O(4)-Na(1)#6	2.715(3)
Na(1)-Rb(1)#12	3.8039(11)	O(4)-Rb(1)#12	3.0717(18)
Na(1)-Rb(1)#13	3.8039(11)	Cl(1)-Rb(1)#18	3.2373(3)
Na(1)-Rb(1)#7	3.9884(16)	Cl(1)-Rb(1)#1	3.2373(3)
N(1)-O(4)#5	1.246(2)	Cl(1)-Rb(1)#3	3.2373(3)
N(1)-O(4)	1.246(2)	Cl(1)-Rb(1)#17	3.2373(3)
N(1)-O(4)#6	1.246(2)	Cl(1)-Rb(1)#11	3.2373(3)
N(1)-Na(1)#5	3.0344(7)		

Symmetry transformations used to generate equivalent atoms:
#1 x-y,x,-z #2 y,x,z #3 -y,x-y,z #4 -x+y,-x+1,z
#5 -y+1,x-y,z #6 -x+y+1,-x+1,-z+1/2 #7 -y+1,x-y,-z+1/2
#8 x-y,-y,-z+1/2 #9 -x+1,-x+y,z #10 y,x-1,z
#11 -x+y,-x,z #12 x,y,-z+1/2 #13 -x+y,-x,-z+1/2
#14 -y,x-y,-z+1/2 #15 -x+y+1,-x+1,z #16 -y+1,x-y+1,z
#17 y,-x+y,-z #18 -x,-y,-z

Table S6. Hydrogen coordinates (× 10^4) and isotropic displacement parameters (Å^2 × 10^3) for $Na_3Rb_6(CO_3)_3(NO_3)_2Br·6H_2O$.

	x	y	z	U_{eq}
H(1)	6456	5613	705	45

Table S7. Hydrogen coordinates (× 10^4) and isotropic displacement parameters (Å^2 × 10^3) for $Na_3Rb_6(CO_3)_3(NO_3)_2Cl·6H_2O$.

	x	y	z	U_{eq}
H(1)	6419	5613	704	48

Table S8. The refractive indices and birefringence values of $Na_3Rb_6(CO_3)_3(NO_3)_2X·6H_2O$ (X=Br and Cl).

Compound		N_o	N_e	Δn
$Na_3Rb_6(CO_3)_3(NO_3)_2Br·6H_2O$	Origin	1.650	1.485	0.165
	CO_3	1.267	1.178	0.089
	NO_3	1.223	1.103	0.121
$Na_3Rb_6(CO_3)_3(NO_3)_2Cl·6H_2O$	Origin	1.642	1.476	0.166
	CO_3	1.266	1.176	0.090
	NO_3	1.223	1.102	0.121

Table S9. Birefringences and band gaps of selected carbonates and nitrates.

	Compound	Birefringence	Band gap (eV)	Reference
1	$Pb_6O_4(BO_3)(NO_3)$	0.276	3.3	[1]
2	$KPb_2(CO_3)_2F$	0.274	3.9	[2]
3	$In(IO_3)_2(NO_3)$	0.269	4.08	[3]
4	$La(OH)_2NO_3$	0.203	4.76	[4]
5	$CsPbCO_3F$	0.183	4.15	[5]
6	$Pb_2(BO_3)(NO_3)$	0.174	3.65	[6]
7	$NaZnCO_3F$	0.171	4.61	[7]
8	$Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$	0.166	4.90	This work
9	$Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$	0.165	4.80	This work
10	$Cs_2Pb(NO_3)_2Br_2$	0.147	3.01	[8]
11	$LiZn(OH)CO_3$	0.147	6.53	[9]
12	$Na_4Zn(CO_3)_3$	0.138	4.06	[7]
13	$RbMgCO_3F$	0.129	6.75	[10]
14	$RbCdCO_3F$	0.121	5.35	[11]
15	$Sr_3[SnOSe_3][CO_3]$	0.12	3.46	[12]
16	$RbCaCO_3F$	0.115	6.22	[13]
17	$NaZnCO_3(OH)$	0.114	6.02	[14]
18	$LiKCO_3$	0.11	6.32	[15]
19	$KCdCO_3F$	0.11	5.46	[16]
20	$Cs_3VO(O_2)_2CO_3$	0.105	2.81	[17]
21	$KY(CO_3)_2$	0.105	6.22	[18]
22	$Lu_8O(CO_3)_3(OH)_{15}Br$	0.088	4.38	[19]
23	$Zn(NH_3)CO_3$	0.087	6.08	[20]
24	$Na_2Gd(CO_3)F_3$	0.084	6.22	[21]
25	$Ca_2Na_3(CO_3)_3F$	0.082	6.52	[22]
26	$Ba_2NO_3(OH)_3$	0.08	6.52	[23]
27	$Ba_3(BO_3)(CO_3)F$	0.048	5.88	[24]
28	$(NH_4)_2Ca_2Y_4(CO_3)_9{\cdot}H_2O$	0.04	6.22	[18]
29	$Cs_3B_8O_{13}(NO_3)$	0.036	6.13	[25]
30	$Ba_2B_5O_8(OH)_2(NO_3){\cdot}3H_2O$	0.033	5.06	[26]
31	$Na_{10}Cd(NO_3)_4(SO_3S)_4$	0.01	3.74	[27]

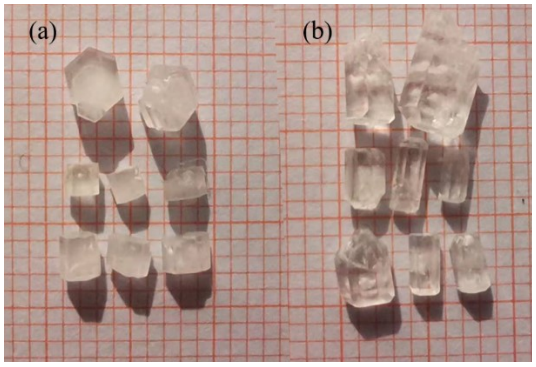

Figure S1. The photos of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$ crystals (the minimum scale is 1 mm).

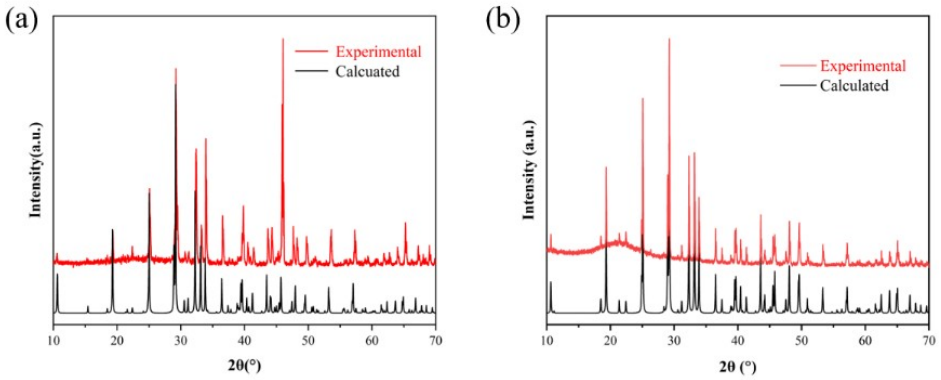

Figure S2. Experimental and theoretical PXRD patterns of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$.

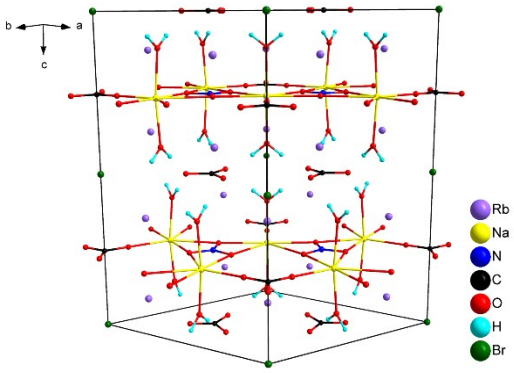

Figure S3. The unit cell of $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$.

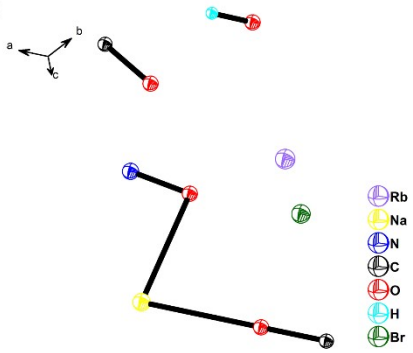

Figure S4. The asymmetric unit $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$.

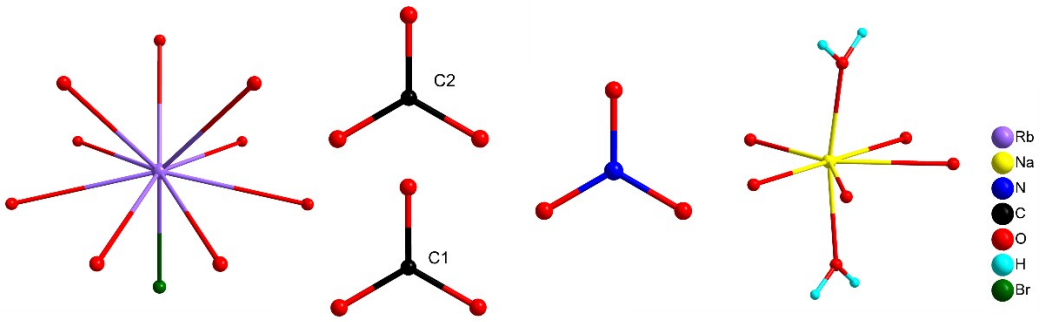

Figure S5. Coordination environments of cations in $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$.

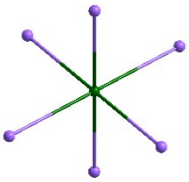

Figure S6. A $[BrRb_6]^{5+}$ polyhedron in $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$.

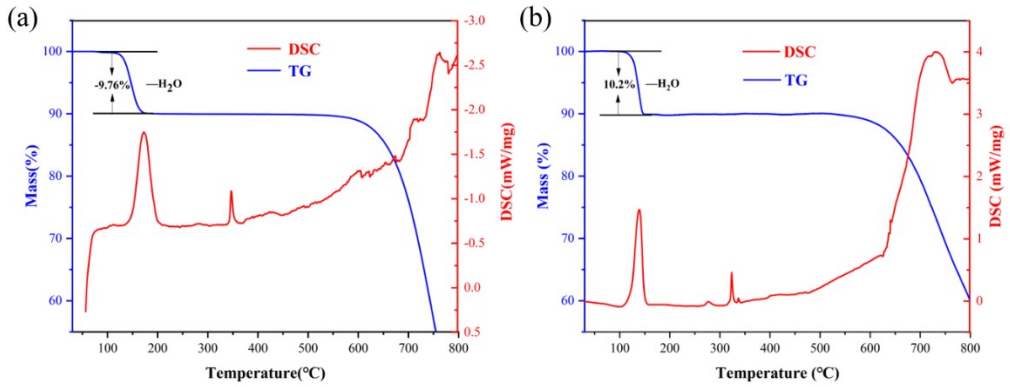

Figure S7. TG-DSC curves of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$.

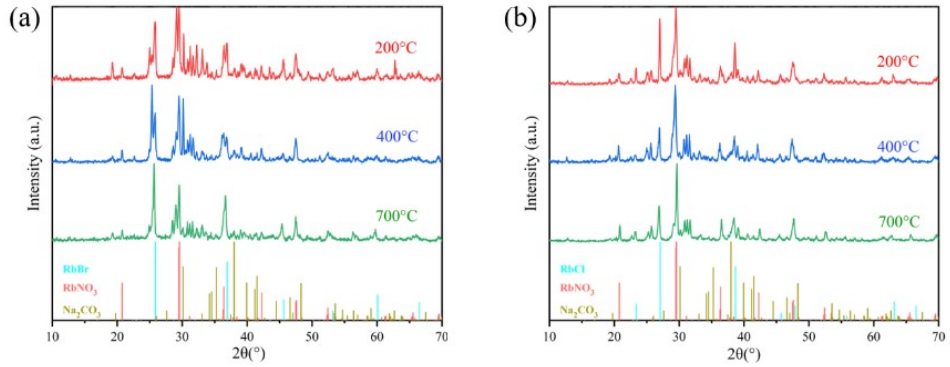

Figure S8. PXRD patterns of residues after decomposition for (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$.

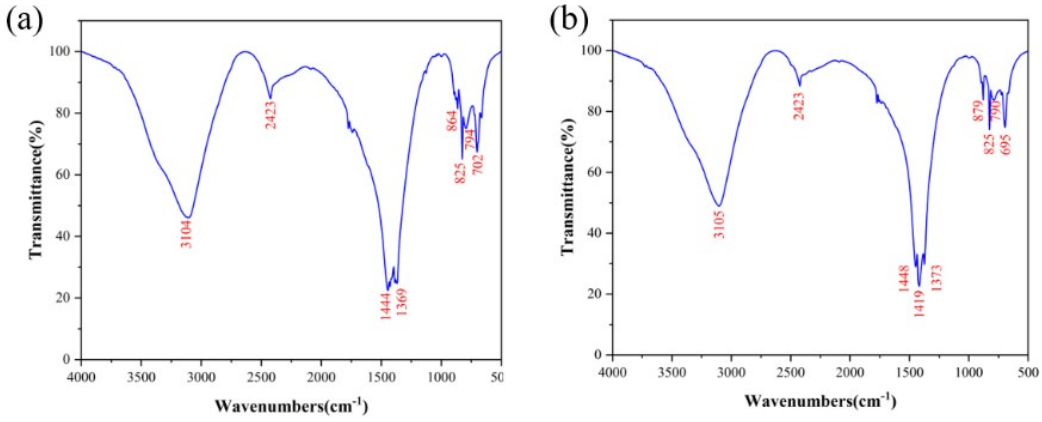

Figure S9. IR spectra of (a) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (b) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$.

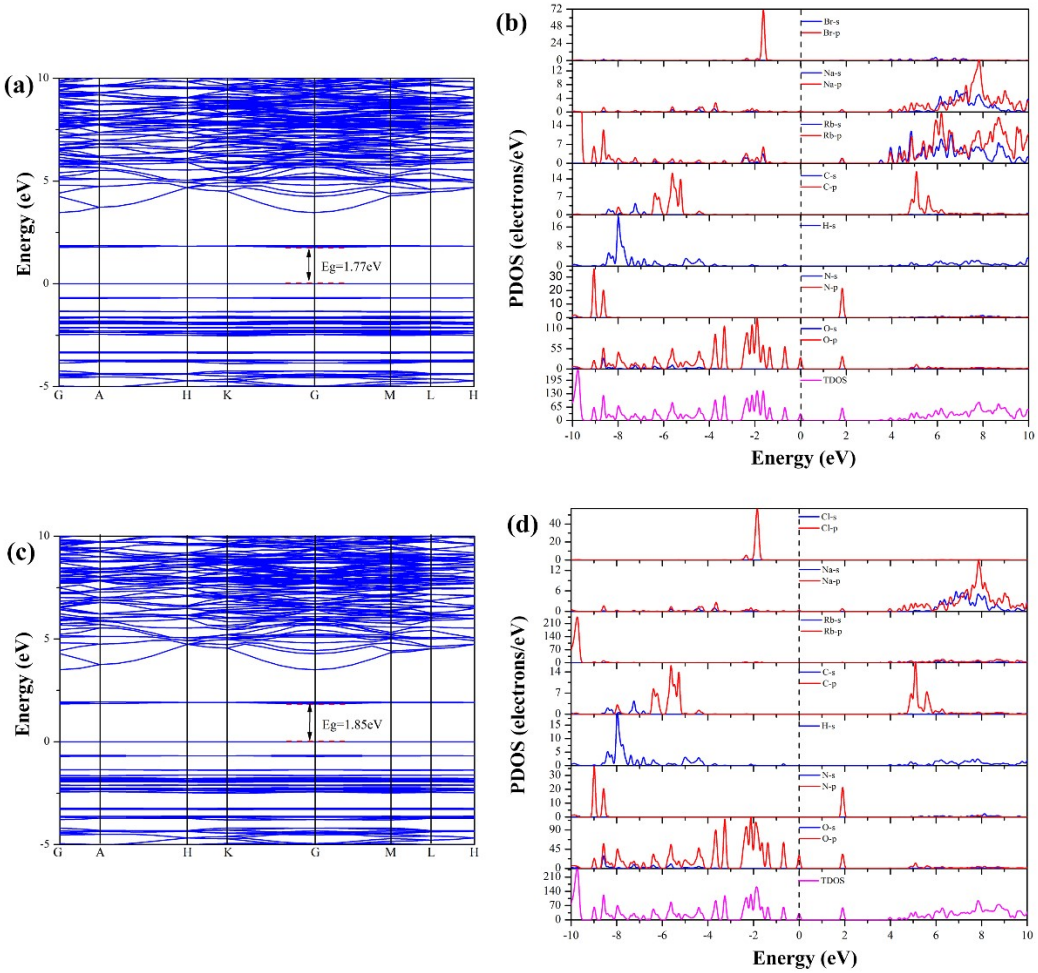

Figure S10. The calculated band gaps and PDOS plots for (a, b) $Na_3Rb_6(CO_3)_3(NO_3)_2Br{\cdot}6H_2O$ and (c, d) $Na_3Rb_6(CO_3)_3(NO_3)_2Cl{\cdot}6H_2O$.

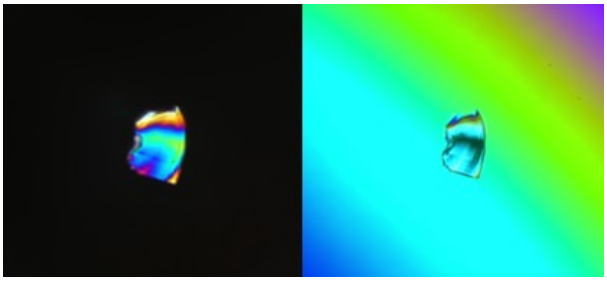

Figure S11. The selected crystal before and after compensatory rotation for birefringence test.

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