Supporting Information

Tunable luminescence based on structural regulation in Organic Antimony Halides for X-ray Scintillation

Sijun Cao [a], Yijia Wang [a], Jun'an Lai [a *], Kang An [b], Mengyue Wu [c], Peng Feng [a], Peng He [a *] and Xiaosheng Tang [c *]

[a] Key Laboratory of Optoelectronic Technology & Systems (Ministry of Education), College of Optoelectronic Engineering, Chongqing University, Chongqing 400044, China

[b] ICT NDT Engineering Research Center, Ministry of Education, Chongqing University, Chongqing 400044, China.

[c] College of Optoelectronic Engineering, Chongqing University of Posts and Telecommunications, Chongqing, 400065, China

* Corresponding author
E-mail: ja.lai@cqu.edu.cn (J. Lai), penghe@cqu.edu.cn (P. He), xstang@cqu.edu.cn (X. Tang).

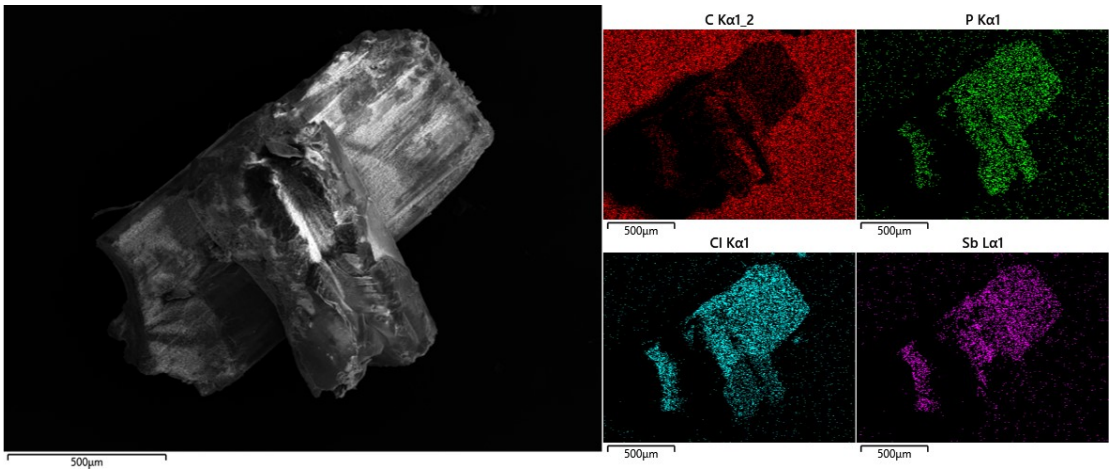

Fig.S1 Scanning electron microscope images of $(NYP)_2SbCl_5$ crystals. EDS mapping images of carbon, phosphorus, chlorine and antimony.

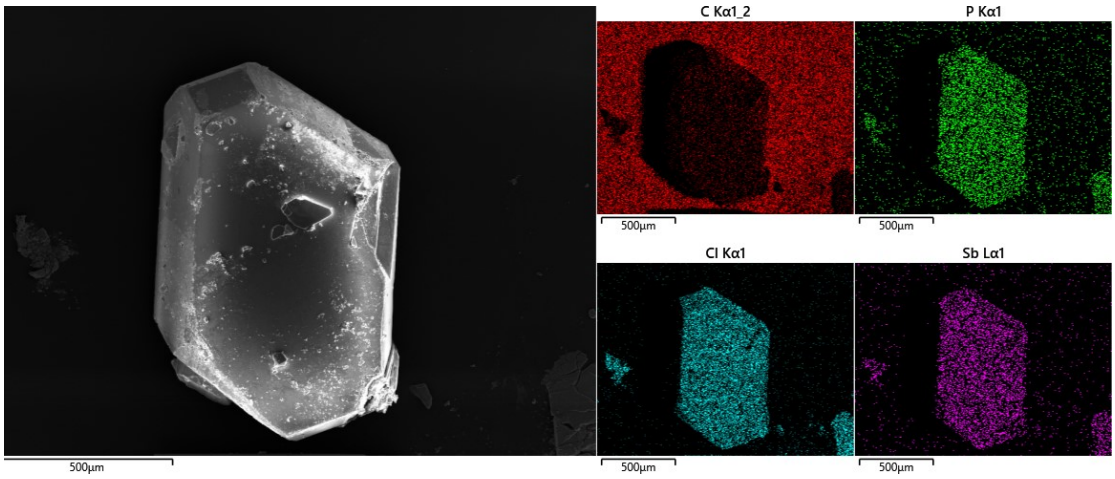

Fig.S2 Scanning electron microscope images of $(NYP)_2Sb_2Cl_8$ crystals. EDS mapping images of carbon, phosphorus, chlorine and antimony.

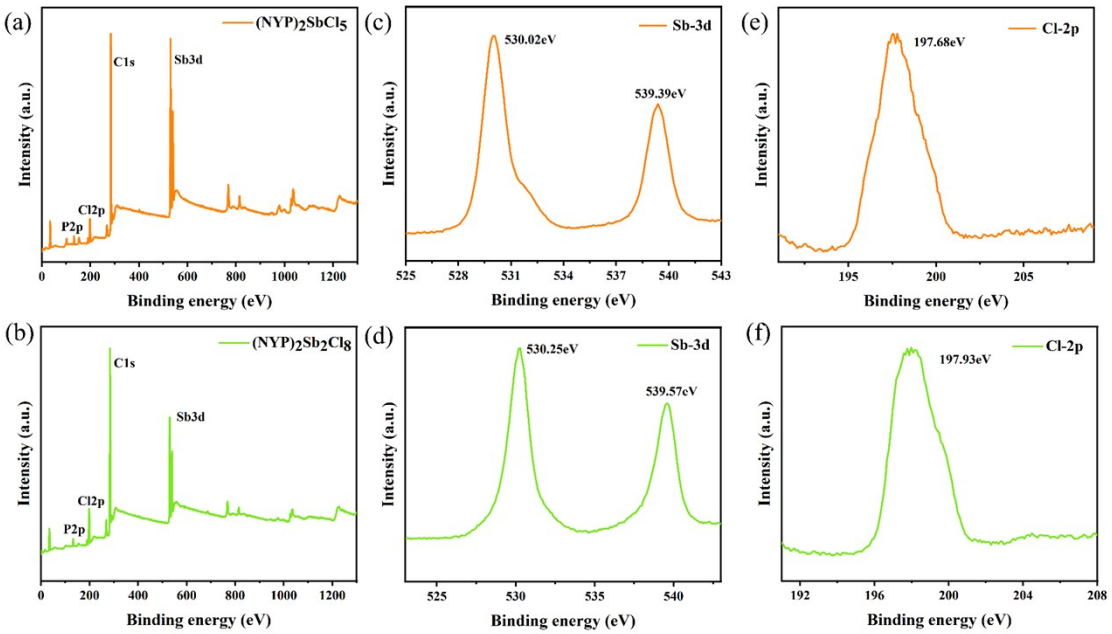

Fig.S3 The XPS spectra of (a, b) all-scan, (c, d) Sb-3d, (e, f) Cl-2p.

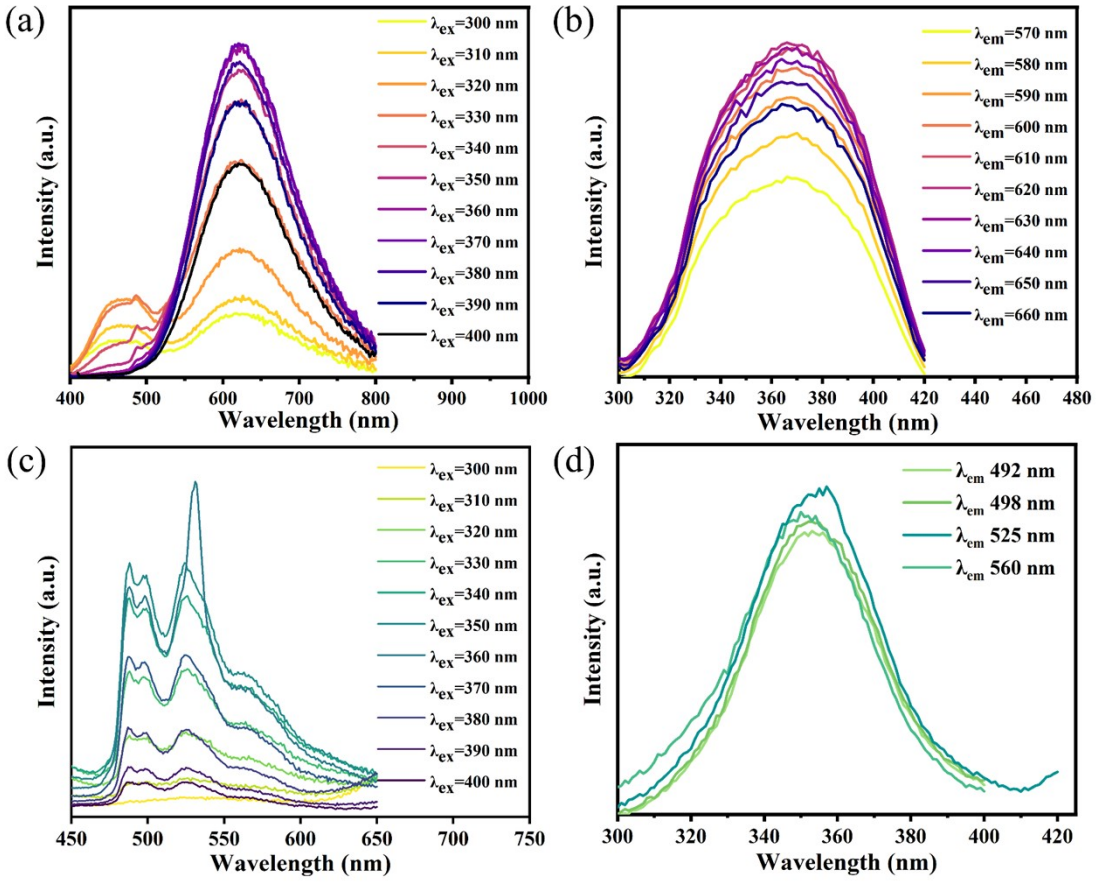

Fig.S4 Emission spectra (a) and excitation spectra (b) of $(NYP)_2SbCl_5$ at different excitation wavelengths. Emission spectra (c) and Excitation spectra (d) of $(NYP)_2Sb_2Cl_8$ at different excitation wavelengths.

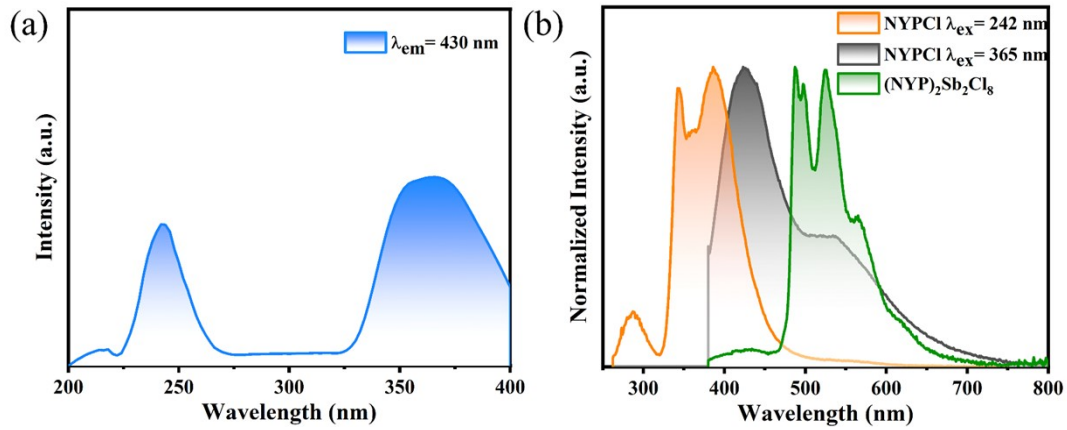

Fig.S5 (a) The PL excitation spectrum of NYPCl with an emission wavelength of 430 nm. (b) The PL spectrums of NYPCl (Excitation wavelengths of 242 and 365 nm) and $(NYP)_2Sb_2Cl_8$.

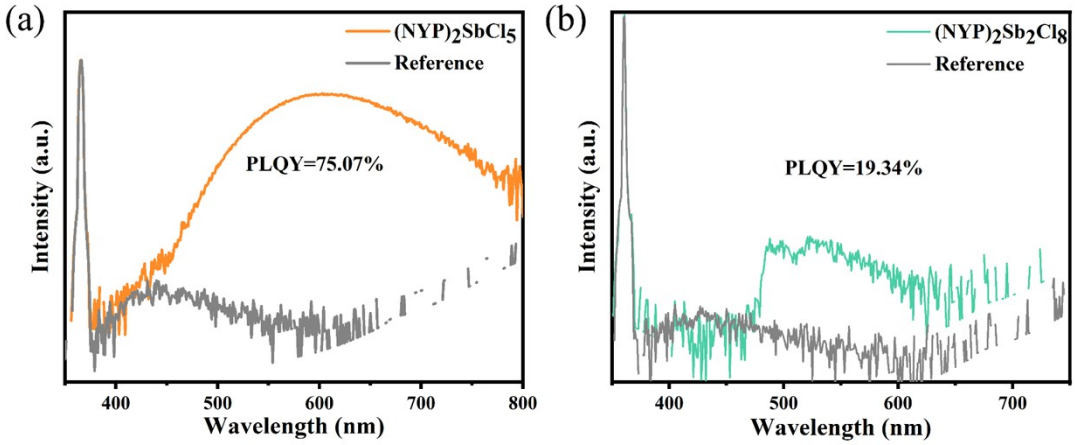

Fig.S6 The PLQY of (a) $(NYP)_2SbCl_5$ (λ_{ex} = 365 nm) and (b) $(NYP)_2Sb_2Cl_8$ (λ_{ex} = 355 nm).

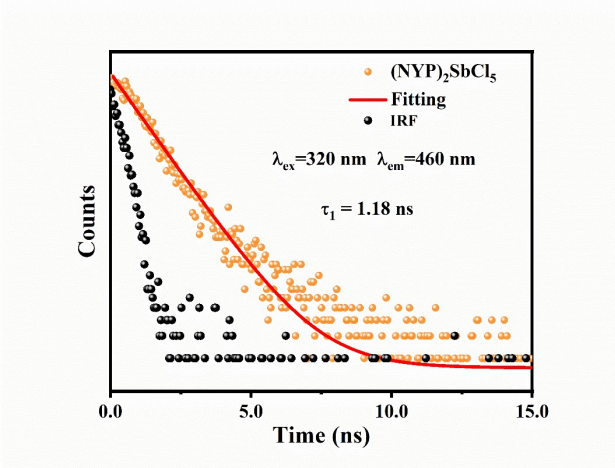

Fig.S7 The time decay curves of $(NYP)_2SbCl_5$ at 320 nm excitation, 460 nm emission.

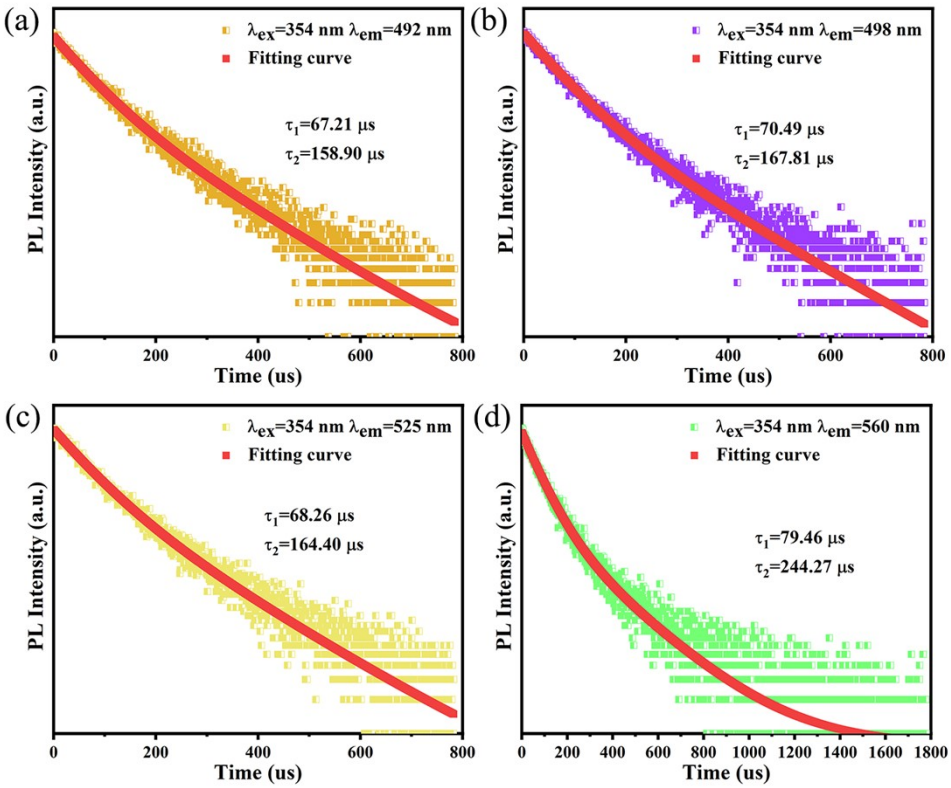

Fig.S8 (a-d) The time decay curves of $(NYP)_2Sb_2Cl_8$ at 354 nm excitation with different emissions.

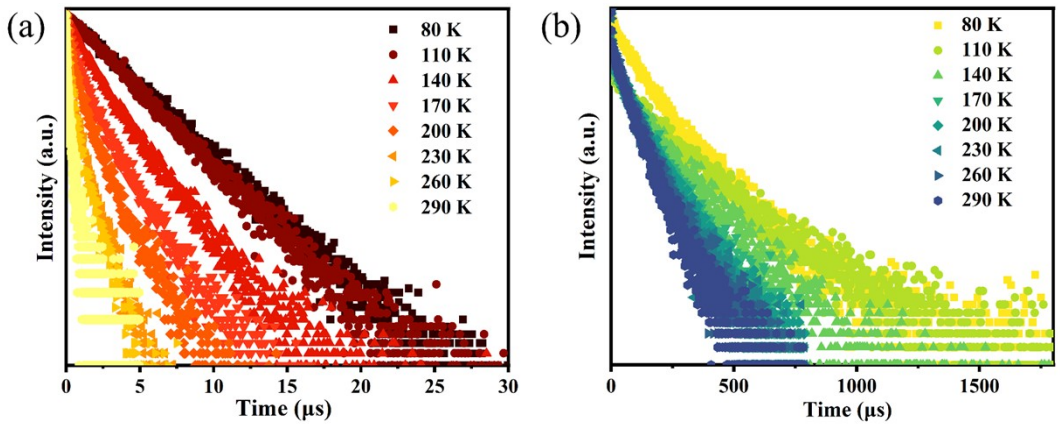

Fig.S9 Temperature-dependent decay curves of (a) $(NYP)_2SbCl_5$ and (b) $(NYP)_2Sb_2Cl_8$ from 80 to 290 K.

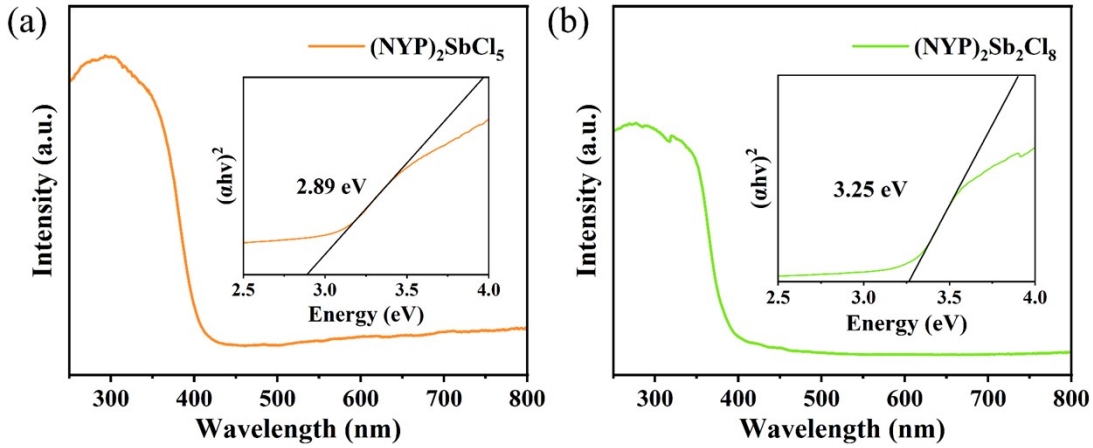

Fig.S10 The Absorption spectra of $(NYP)_2SbCl_5$ and $(NYP)_2Sb_2Cl_8$.

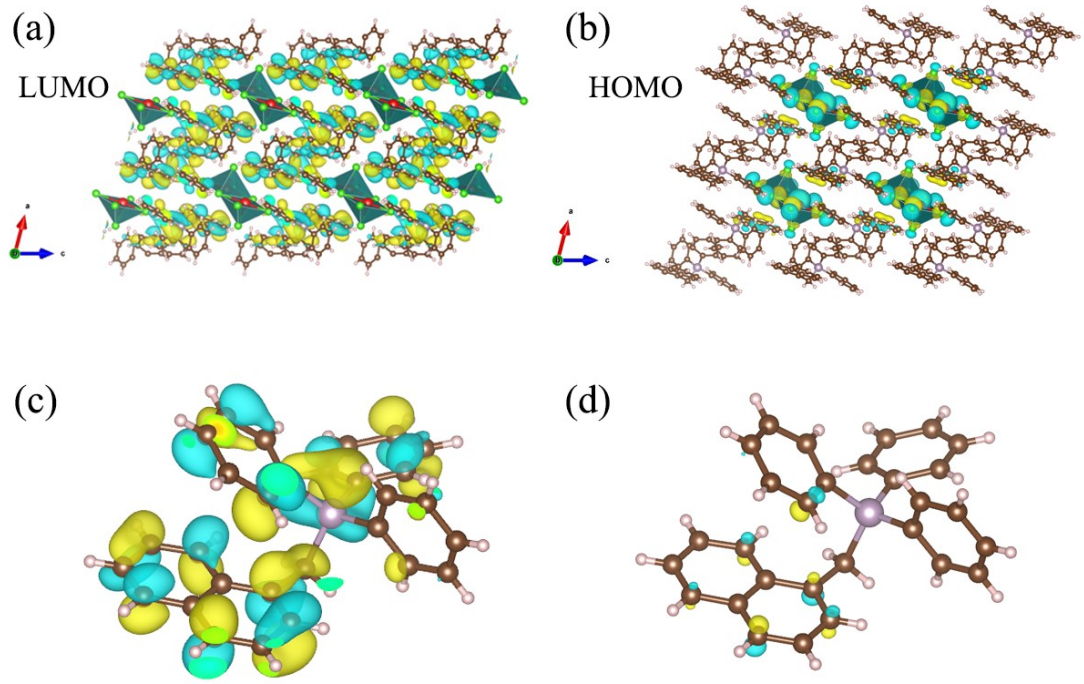

Fig.S11 Isosurface plots of charge density corresponding to (a) LUMO and (b) HOMO of $(NYP)_2Sb_2Cl_8$. Charge distribution maps of NYP at (c) low and (d) high isosurface levels.

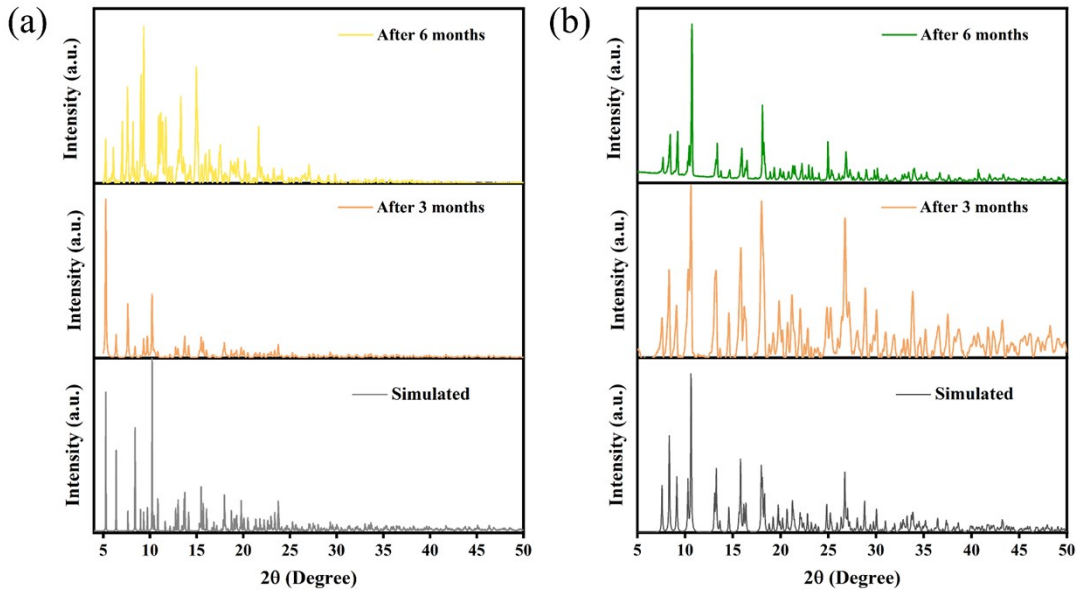

Fig.S12 The PXRD spectra of (a) $(NYP)_2SbCl_5$ and (b) $(NYP)_2Sb_2Cl_8$ after

3 and 6 months of storage in ambient atmosphere.

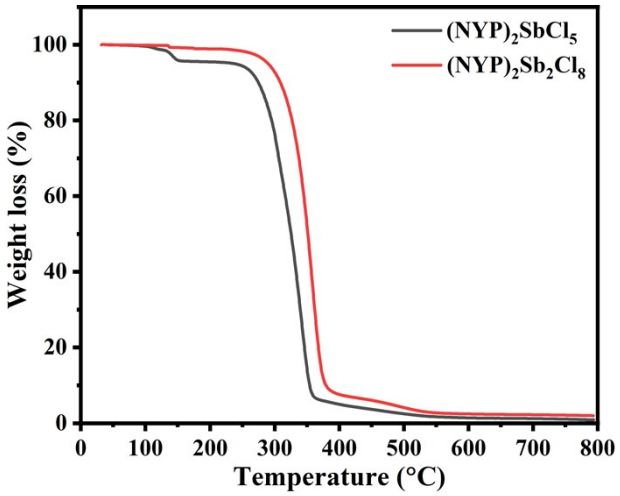

Fig.S13 The thermogravimetry analysis of $(NYP)_2SbCl_5$ and $(NYP)_2Sb_2Cl_8$.

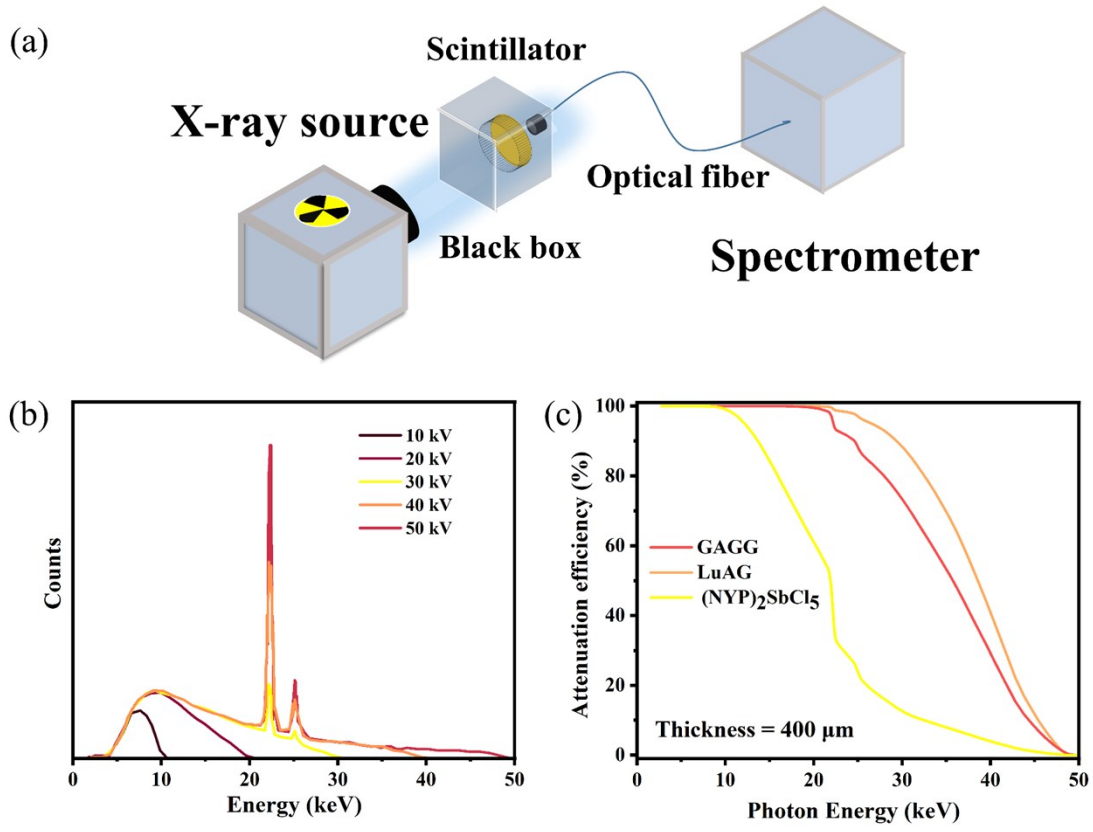

Fig.S14 (a) Schematic of the RL intensity measurement testing system. (b) Output spectra at different tube voltages of Mini-X X-ray tube (target: Ag, the characteristic peak is at 22 keV). (c) Calculated X-ray attenuation efficiency spectra of GAGG, LuAG, and $(NYP)_2SbCl_5$ at the thickness of 400 μm.

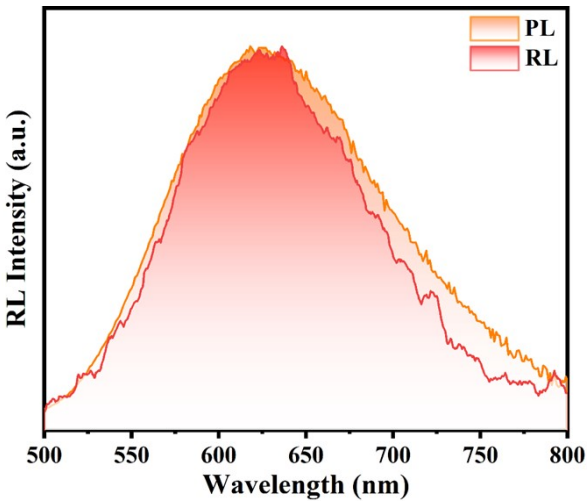

Fig.S15 The PL and RL of $(NYP)_2SbCl_5$.

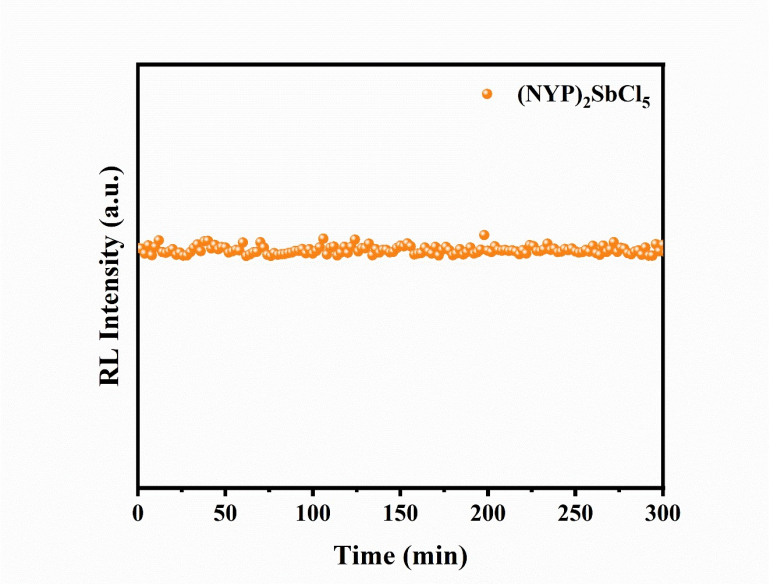

Fig.S16 The radiation stability of $(NYP)_2SbCl_5$.

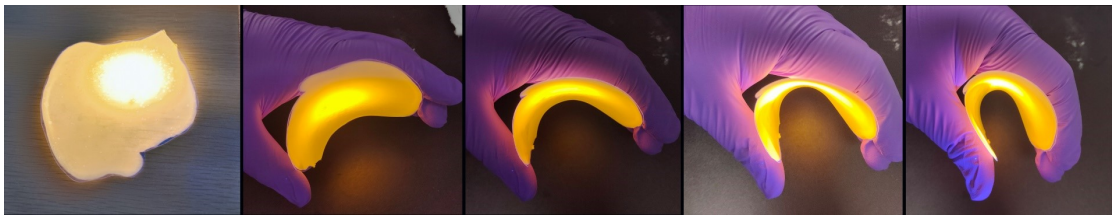

Fig.S17 The UV-emitting pictures of scintillating screens at different bending angles.

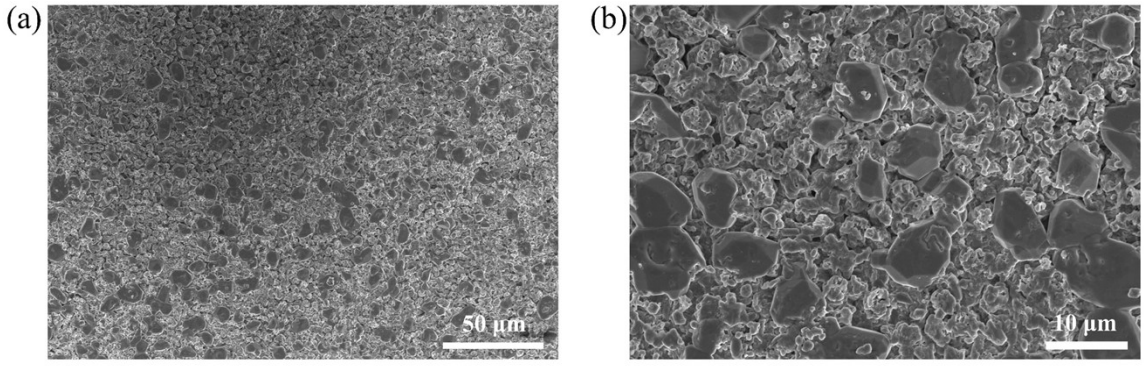

Fig.S18 (a) SEM and (a) detailed images of the scintillating film based on $(NYP)_2SbCl_5$.

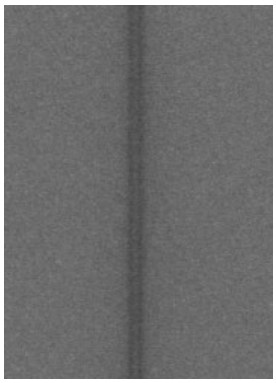

Fig.S19 The imaging picture of the D12 (7.7 lp/mm) line pair extracted from the line pair card.

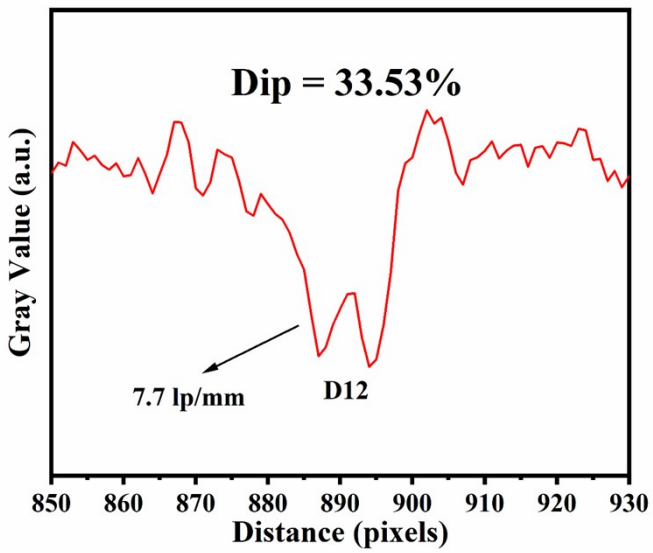

Fig.S20 Gray value profiles of the line pair D12 (7.7 lp/mm) extracted from the line pair card.

Table S1. Crystal data and structure refinement for $(NYP)_2SbCl_5$.

Empirical formula	C58 H48 Cl5 P2 Sb
Formula weight	1105.90
Temperature	296.15 K
Wavelength	0.71073 Å
Crystal system	Orthorhombic
Space group	P21
Unit cell dimensions	a = 10.0922(19) Å a = 90 °. b= 21.053(4) Å b = 90 °. c = 27.752(6) Å g = 90 °.
Volume	5896(2) $Å^3$
Z	4
Density (calculated)	1.246 g/cm^3
Absorption coefficient	0.782 mm^{-1}
F(000)	2248.0
Theta range for data collection	2.542 to 25.019°.
Index ranges	-12<=h<=12, -25<=k<=21, -32<=l<=32
Reflections collected	92143
Independent reflections	10367 [R(int) = 0.1035]
Completeness to theta = 25.115°	99.8 %
Data / restraints / parameters	10367 / 1193 / 782
Goodness-of-fit on F^2	1.040
Final R indices [I>2sigma(I)]	R1 = 0.0542, wR2 = 0.1094
R indices (all data)	R1 = 0.1082, wR2 = 0.1294
Largest diff. peak and hole	0.613 and -0.375 $e.Å^{-3}$

Table S2. Bond Lengths for $(NYP)_2SbCl_5$.

Atom	Length/Å	Atom	Length/Å	Atom	Length/Å	Atom	Length/Å
Sb(01)-Cl(03)	2.629(3)	C(015)-H(015)	0.93	C(01B)-H(01B)	0.93	C(15A)-H(15A)	0.93
Sb(01)-Cl(04)	2.631(3)	C(015)-C(01F)	1.39	C(01C)-H(01E)	0.93	C(15A)-C(14A)	1.39
Sb(01)-Cl(05)	2.368(3)	C(01F)-H(01F)	0.93	C(01C)-C(01R)	1.34(3)	C(14A)-H(14A)	0.93
Sb(01)-Cl(06)	2.585(3)	C(01F)-C(01L)	1.39	C(01H)-H(01A)	0.97	C(14A)-C(13A)	1.39
Sb(01)-Cl(07)	2.592(3)	C(01L)-H(01L)	0.93	C(01H)-H(01C)	0.97	C(13A)-H(13A)	0.93
P(002)-C(009)	1.781(10)	C(01L)-C(01I)	1.39	C(01H)-C(1B)	1.57(3)	C(13A)-C(10A)	1.39
P(002)-C(00H)	1.783(11)	C(01I)-C(01A)	1.39	C(01R)-H(01R)	0.93	C(10A)-H(10A)	0.93
P(002)-C(00J)	1.819(9)	C(01I)-C(01C)	1.47(3)	C(01R)-C(39)	1.29(3)	C(10A)-C(01E)	1.39
P(002)-C(00U)	1.790(10)	C(01A)-C(00W)	1.45(4)	C(1)-H(1)	0.93	C(01E)-H(01H)	0.93
P(008)-C(00Q)	1.705(18)	C(00P)-H(00P)	0.93	C(2)-H(2)	0.93	C(0)-H(0)	0.93
P(008)-C(01H)	1.81(2)	C(00P)-C(010)	1.343(14)	C(2)-C(3)	1.39	C(0)-C(17)	1.44(3)
P(008)-C(00V)	1.768(13)	C(00Q)-C(01M)	1.39	C(2)-C(00V)	1.39	C(11)-H(11)	0.93
P(008)-C(16)	1.813(14)	C(00Q)-C(018)	1.39	C(3)-H(3)	0.93	C(11)-C(18)	1.33(3)
C(009)-C(00F)	1.405(14)	C(01M)-H(01M)	0.93	C(3)-C(01N)	1.39	C(12)-H(12)	0.93
C(009)-C(1)	1.409(14)	C(01M)-C(013)	1.39	C(01N)-H(01N)	0.93	C(12)-C(19)	1.39
C(00A)-C(00B)	1.441(14)	C(013)-H(013)	0.93	C(01N)-C(4)	1.39	C(12)-C(1A)	1.39
C(00A)-C(00I)	1.405(16)	C(013)-C(00S)	1.39	C(4)-H(4)	0.93	C(19)-H(19)	0.93
C(00A)-C(00Y)	1.385(15)	C(00S)-H(00S)	0.93	C(4)-C(5)	1.39	C(19)-C(01G)	1.39
C(00B)-C(00E)	1.428(14)	C(00S)-C(019)	1.39	C(5)-H(5)	0.93	C(01G)-H(01G)	0.93
C(00B)-C(00K)	1.407(14)	C(019)-H(019)	0.93	C(5)-C(00V)	1.39	C(01G)-C(1D)	1.39
C(00D)-H(00D)	0.93	C(019)-C(018)	1.39	C(16)-C(15)	1.39	C(1D)-H(1D)	0.93
C(00D)-C(00E)	1.366(14)	C(018)-H(018)	0.93	C(16)-C(01D)	1.39	C(1D)-C(01J)	1.39
C(00D)-C(00M)	1.415(16)	C(00R)-H(00R)	0.93	C(15)-H(15)	0.93	C(01J)-C(1A)	1.39
C(00E)-C(00J)	1.462(12)	C(00R)-C(00X)	1.341(16)	C(15)-C(14)	1.39	C(1A)-C(1B)	1.35(3)
C(00F)-H(00F)	0.93	C(00T)-H(00T)	0.93	C(14)-H(14)	0.93	C(2A)-H(2A)	0.93
C(00F)-C(010)	1.387(14)	C(00T)-C(016)	1.380(16)	C(14)-C(13)	1.39	C(2A)-C(3A)	1.39
C(00G)-H(00G)	0.93	C(00U)-C(00Z)	1.365(16)	C(13)-H(13)	0.93	C(2A)-C(9)	1.39
C(00G)-C(00P)	1.381(15)	C(00U)-C(017)	1.387(15)	C(13)-C(10)	1.39	C(3A)-H(3A)	0.93
C(00G)-C(1)	1.403(14)	C(00W)-H(00W)	0.93	C(10)-H(10)	0.93	C(3A)-C(01O)	1.39
C(00H)-C(00N)	1.408(14)	C(00W)-C(39)	1.54(5)	C(10)-C(01D)	1.39	C(01O)-H(01O)	0.93
C(00H)-C(012)	1.378(14)	C(00X)-H(00X)	0.93	C(01D)-H(01D)	0.93	C(01O)-C(4A)	1.39
C(00I)-H(00I)	0.93	C(00X)-C(012)	1.397(16)	C(39)-H(39)	0.93	C(4A)-H(4A)	0.93
C(00I)-C(00M)	1.360(17)	C(00Y)-H(00Y)	0.93	C(6)-H(6)	0.93	C(4A)-C(5A)	1.39
C(00J)-H(00A)	0.97	C(00Y)-C(016)	1.340(16)	C(6)-C(01J)	1.38(3)	C(5A)-H(5A)	0.93
C(00J)-H(00B)	0.97	C(00Z)-H(00Z)	0.93	C(6)-C(01S)	1.35(4)	C(5A)-C(9)	1.39
C(00K)-H(00K)	0.93	C(00Z)-C(011)	1.418(15)	P(0)-C(7)	1.80(3)	C(17)-H(17)	0.93
C(00K)-C(00T)	1.360(14)	C(010)-H(010)	0.93	P(0)-C(8)	1.81(2)	C(17)-C(1C)	1.32(3)
C(00L)-H(00L)	0.93	C(011)-H(011)	0.93	P(0)-C(16A)	1.761(14)	C(18)-H(18)	0.93
C(00L)-C(00N)	1.365(15)	C(011)-C(014)	1.33(2)	P(0)-C(9)	1.771(13)	C(18)-C(1C)	1.36(4)
C(00L)-C(00R)	1.371(16)	C(012)-H(012)	0.93	C(7)-C(0)	1.43(3)	C(1B)-C(39A)	1.48(5)
C(00M)-H(00M)	0.93	C(014)-H(014)	0.93	C(7)-C(11)	1.36(3)	C(1C)-H(1C)	0.93

C(00N)-H(00N)	0.93	C(014)-C(01B)	1.38(2)	C(8)-H(8A)	0.97	C(39A)-H(39A)	0.93
C(00O)-C(015)	1.39	C(016)-H(016)	0.93	C(8)-H(8B)	0.97	C(39A)-C(01S)	1.29(4)
C(00O)-C(01A)	1.39	C(017)-H(017)	0.93	C(16A)-C(15A)	1.39	C(01S)-H(01S)	0.93
C(00O)-C(8)	1.46(2)	C(017)-C(01B)	1.412(19)	C(16A)-C(01E)	1.39		

Table S3. Crystal data and structure refinement for $(NYP)_2Sb_2Cl_8$.

Empirical formula	C29 H24 Cl4 P Sb
Formula weight	667.00
Temperature	300.00 K
Crystal system	triclinic
Space group	P_{-1}
Unit cell dimensions	a = 10.843(4) Å a = 63.782(10) °. b= 12.238(5) Å b = 67.746(11) °. c = 13.423(5) Å g = 67.259(13) °.
Volume	1426.5(10) $Å^3$
Z	2
Density (calculated)	1.553 g/cm^3
Absorption coefficient	7.970 mm^{-1}
F(000)	664.0
Theta range for data collection	7.946 to 120.47°.
Index ranges	-13<=h<=13, -14<=k<=15, -17<=l<=16
Reflections collected	16695
Independent reflections	6044 [R(int) = 0.0356, R(sigma) = 0.0368]
Completeness to theta = 25.115°	99.4 %
Data / restraints / parameters	6044 / 0 / 317
Goodness-of-fit on F^2	1.135
Final R indices [I>2sigma(I)]	R_1 = 0.0373, wR_2 = 0.0936
R indices (all data)	R_1 = 0.0463, wR_2 = 0.1053
Largest diff. peak and hole	0.80 and -1.28 $e.Å^{-3}$

Table S4. Bond Lengths for $(NYP)_2Sb_2Cl_8$.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
Sb1	Cl2	2.3804(15)	C7	C12	1.400(6)
Sb1	Cl3	2.4806(15)	C7	C8	1.384(6)
Sb1	Cl1	2.4471(15)	C14	C15	1.394(6)
P1	C1	1.805(4)	C12	C11	1.392(6)
P1	C13	1.801(4)	C4	C3	1.382(7)
P1	C19	1.825(4)	C4	C5	1.368(7)
P1	C7	1.792(4)	C28	C27	1.371(6)
C1	C2	1.395(6)	C6	C5	1.392(6)
C1	C6	1.391(5)	C9	C8	1.390(6)
C13	C18	1.402(6)	C9	C10	1.363(7)
C13	C14	1.385(6)	C24	C23	1.423(8)
C20	C29	1.421(5)	C24	C25	1.408(7)
C20	C19	1.516(5)	C10	C11	1.380(7)
C20	C21	1.375(6)	C27	C26	1.398(8)
C29	C28	1.430(6)	C22	C23	1.354(8)
C29	C24	1.427(6)	C17	C16	1.384(8)
C18	C17	1.385(6)	C15	C16	1.365(8)
C21	C22	1.412(7)	C26	C25	1.365(9)
C2	C3	1.390(6)			

Table S5. Summary of the PL properties of hybrid antimony chlorides with $SbCl_5$ cluster.

Compound	Emission (nm)	Decay time (μs)	PLQY (%)	Sb-Sb distance (Å)	Ref
$(PPN)_2SbCl_5$	635	4.1	98.1	12.19	[1]
$(Bmim)_2SbCl_5$	583	4.26	86.3	9.02	[2]
$(TMA)_2SbCl_5{\cdot}DMF$	630	6.62	67.2	8.61	[3]
$(C_6N_2H_{16})_2SbCl_5$	613	1.06	25.3	3.99	[4]
$(C_6N_2H_{16})_2SbCl_5{\cdot}H_2O$	620	1.22	39.6	4.34	[4]
$(NYP)_2SbCl_5$	620	0.33	75.07	10.09	This work

Table S6 The parameter of ISO 19232 duplex-wire image quality indicator. DD: duplex wire numbers; UT: corresponding unsharpness; SRb: corresponding basic spatial resolution; d: wire diameter and spacing; LP: corresponding line pair value.

DD	UT (mm)	SRb (mm)	d (mm)	LP (lp/mm)
D4	800	400	400	1.250
D5	640	320	320	1.560
D6	500	250	250	2.000
D7	400	200	200	2.500
D8	320	160	160	3.130
D9	260	130	130	3.850
D10	200	100	100	5.000
D11	160	80	80	6.250
D12	130	63	63	7.690
D13	100	50	50	10.000
D14	80	40	40	12.50
D15	63	32	32	15.90
D16	50	25	25	20.00
D17	20	20	20	25.00

1. Q. He, C. Zhou, L. Xu, S. Lee, X. Lin, J. Neu, M. Worku, M. Chaaban and B. Ma, Highly Stable Organic Antimony Halide Crystals for X-ray Scintillation, *ACS Mater. Lett.*, 2020, **2**, 633-638.
2. Z.-P. Wang, J.-Y. Wang, J.-R. Li, M.-L. Feng, G.-D. Zou and X.-Y. Huang, [Bmim]2SbCl5: a main group metal-containing ionic liquid exhibiting tunable photoluminescence and white-light emission, *Chem. Commun.*, 2015, **51**, 3094-3097.
3. Q. Wei, T. Chang, R. Zeng, S. Cao, J. Zhao, X. Han, L. Wang and B. Zou, Self-Trapped Exciton Emission in a Zero-Dimensional (TMA)2SbCl5·DMF Single Crystal and Molecular Dynamics Simulation of Structural Stability, *The Journal of Physical Chemistry Letters*, 2021, **12**, 7091-7099.
4. G. Song, M. Li, S. Zhang, N. Wang, P. Gong, Z. Xia and Z. Lin, Enhancing Photoluminescence Quantum Yield in 0D Metal Halides by Introducing Water Molecules, *Adv. Funct. Mater.*, 2020, **30**, 2002468.