Nanoscale Layer-Separated Zincophosphate Framework Enabled by Dual-Mode Aromatic Pillar Engineering for Selective Luminescent Dye Detection

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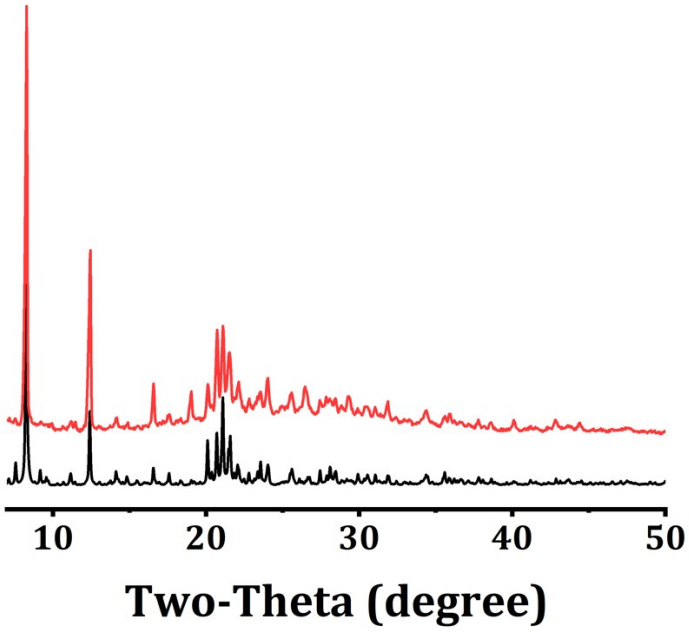

Fig. S1 X-ray powder patterns of **NTOU-11** (red) and the simulated pattern derived from single-crystal X-ray diffraction (black).

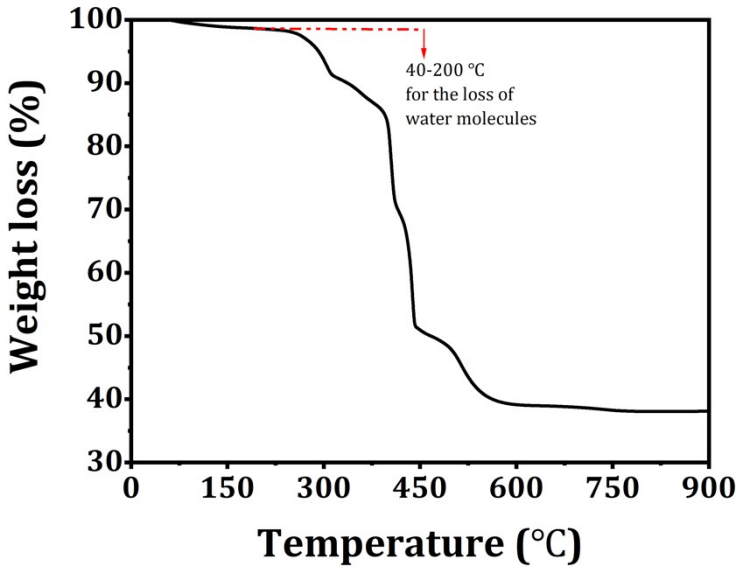

Fig. S2 TGA curve of **NTOU-11** in O_2 at a heating rate of 5°C/ min^{-1} from 40 °C to 900 °C.

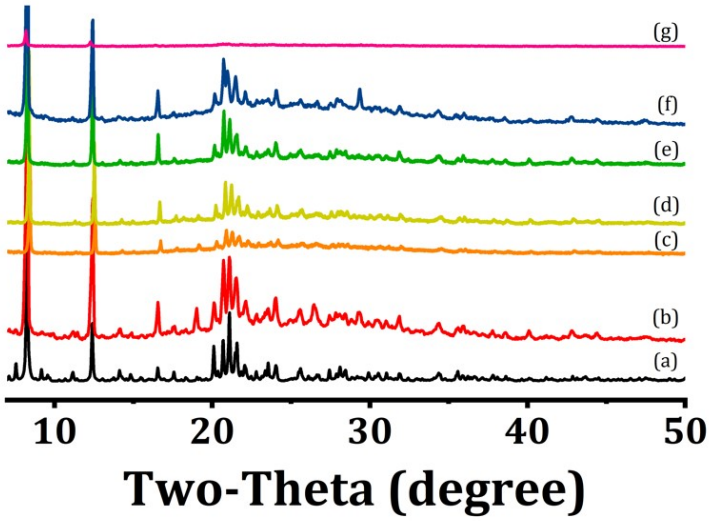

Fig. S3 X-ray powder patterns of **NTOU-11** for thermal stability studies: (a) simulated, (b) as-synthesized, (c) after holding at 100°C for 30 min, (d) 200°C (e) 250°C (f) 300°C (g) 350°C.

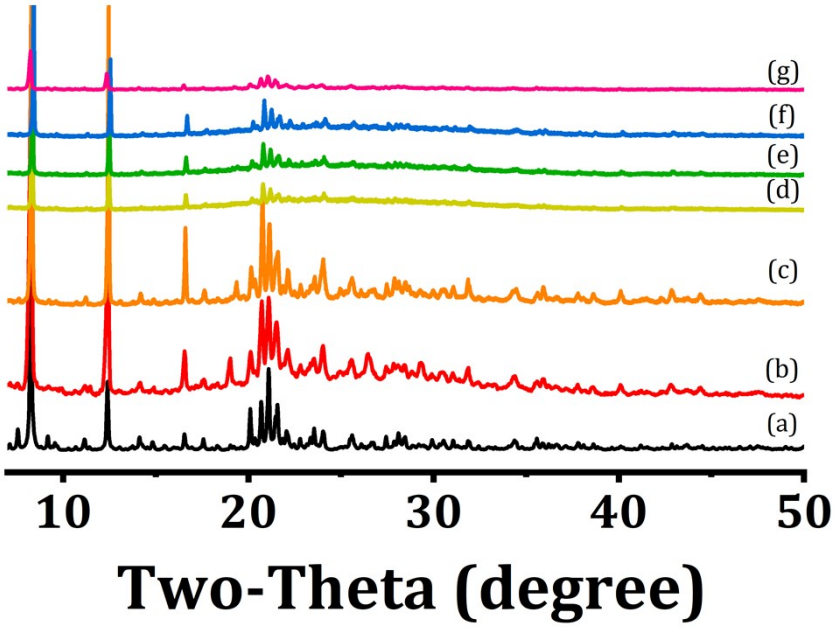

Fig. S4 X-ray powder patterns of **NTOU-11** powder after stirring in aqueous solutions at different pH values for 1 h: (a) simulated, (b) as-synthesized, (c) pH4, (d) pH6, (e) pH8, (f) pH10, (g) pH12.

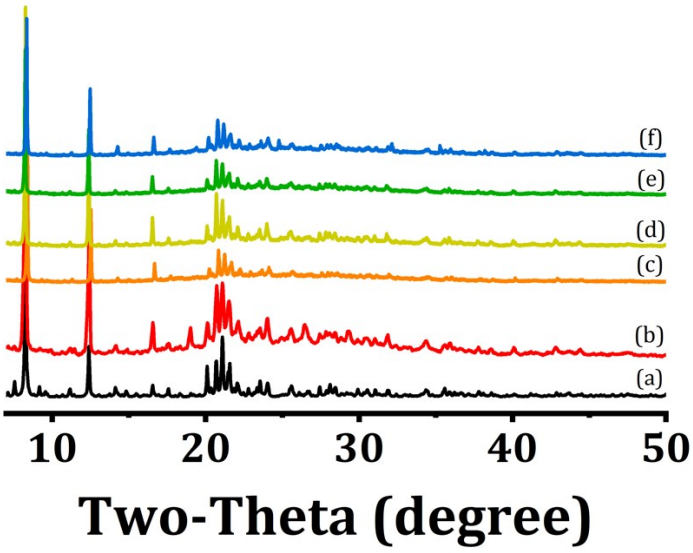

Fig. S5 X-ray powder patterns of **NTOU-11** for chemical stability studies: (a) simulated (b) as-synthesized, after stirring for 7 hours in (c) water, (d) ethanol, (e) DMF, and (f) 0.1 M PBS.

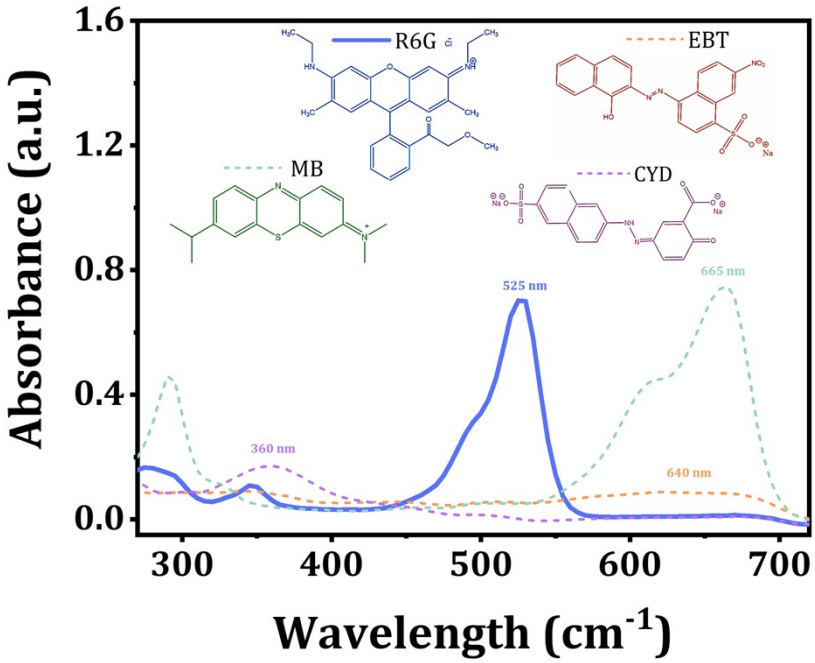

Fig. S6 (a) Absorption peaks of Rhodamine 6G (pink), Eriochrome black T (violet), Methylene blue (blue), and Chrome yellow D (chartreuse). (b) Molecular structures of organic dyes and the wavelength proximity between their absorption peaks and the emission peak of **NTOU-11**.

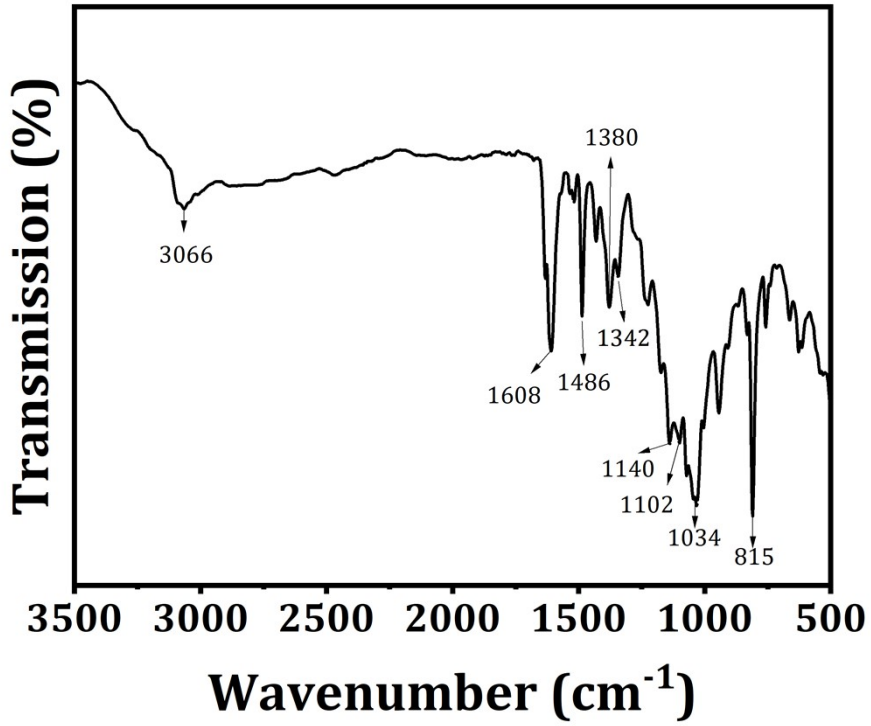

Fig. S7 FTIR spectrum of **NTOU-11**.

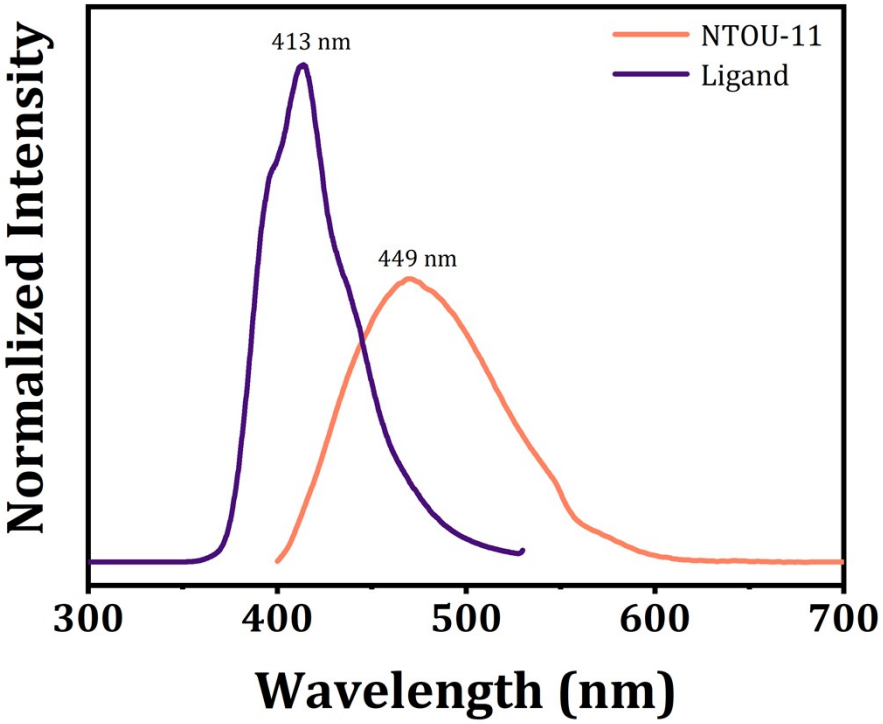

Fig. S8 PL spectra of **NTOU-11** and the DPBP ligand (λ_{ex} = 369 nm).

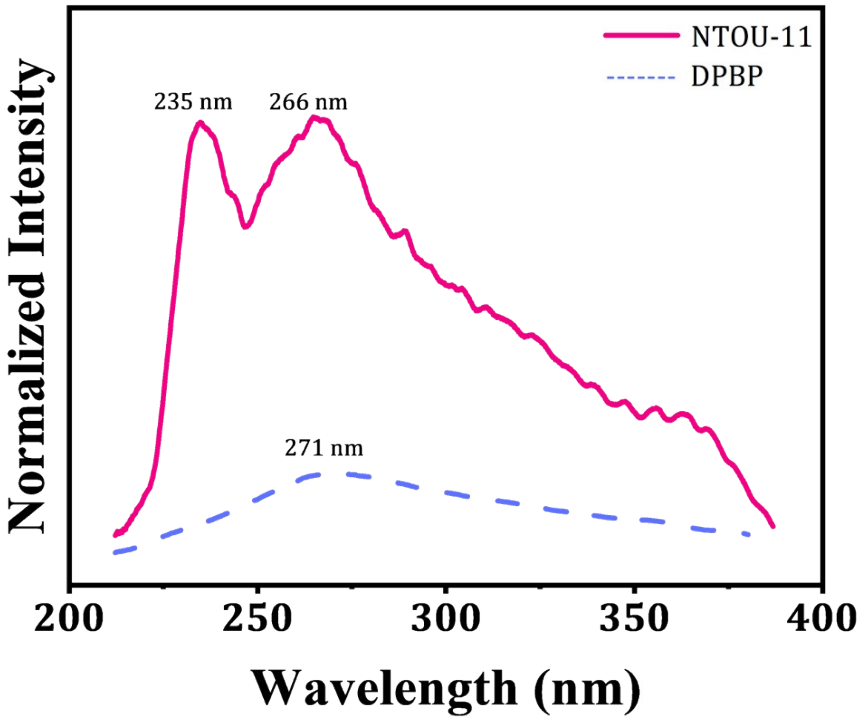

Fig. S9 Excitation spectra of the free DPBP and compound **NTOU-11**.

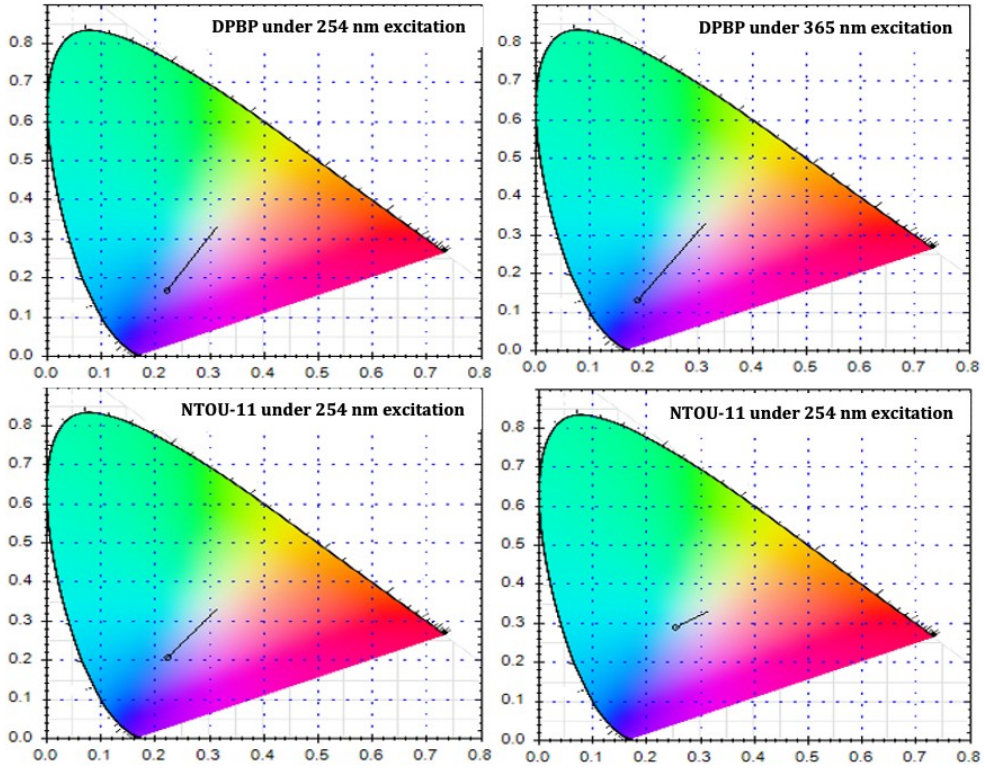

Fig. S10 CIE chromaticity diagrams of the free DPBP ligand (top) and **NTOU-11** (bottom) under UV light irradiation at 254 nm and 365 nm.

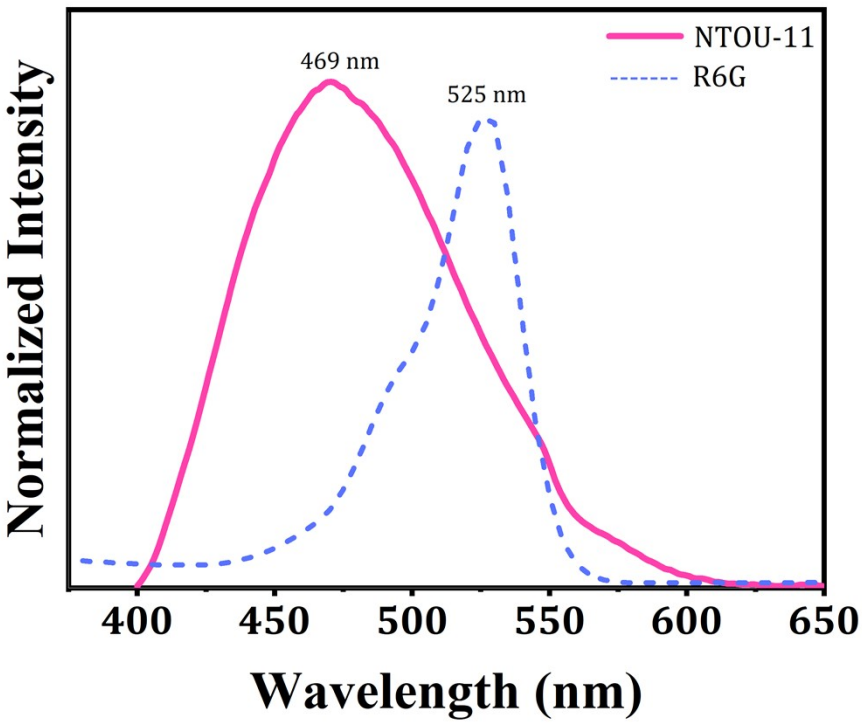

Fig. S11 The absorption peak of Rhodamine 6G and the emission peak of **NTOU-11**.

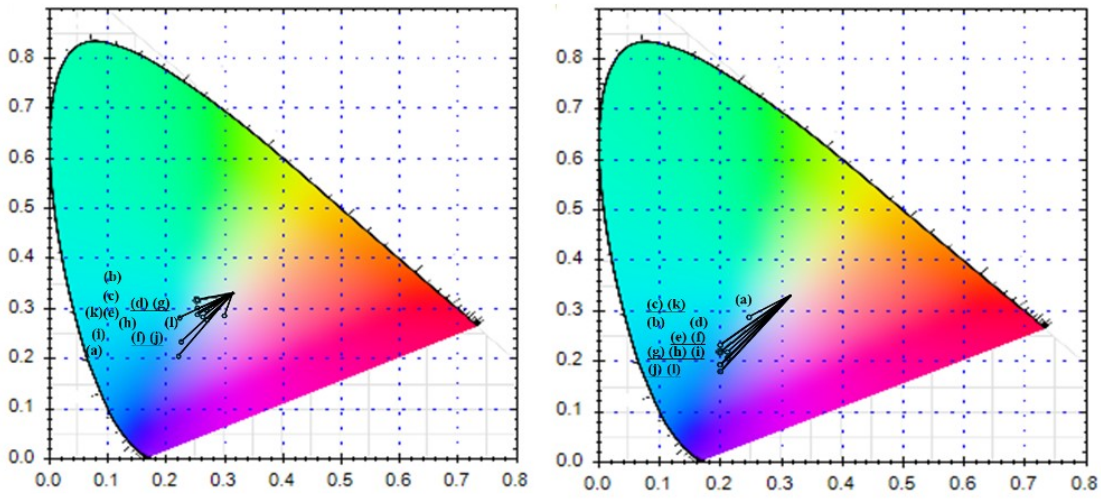

Fig. S12 CIE chromaticity diagrams of **NTOU-11** and **R6G@NTOU-11** powders with concentrations ranging of R6G from 0.001 mM to 20 mM under 254 nm (left) and 365 nm (right) UV light irradiation. (a) compound CY (b) 0.001 mM R6G (c) 0.01 mM R6G (d) 0.05 mM R6G (e) 0.1 mM R6G (f) 0.25 mM R6G (g) 0.5 mM R6G (h) 1 mM R6G (i) 5 mM R6G (j) 10 mM R6G (k) 15 mM R6G (l) 20 mM R6G.

Table S1. Comparison of structural features between NTOU-11 and related materials.

Compound	Coordination Modes of Organic Molecules	Inorganic Motifs	Structural Variations
NTHU-2 $(H_2tmdp)[(ZnHPO_4)_2(BDC)]$	BDC acts as an anionic pillar; tmdp acts as a protonated cation.	Corrugated $[4.8^2]$ inorganic net of alternating ZnO_4 and HPO_4 tetrahedra.	Exhibits a bimodal pore size distribution (1.36 and 21.8 nm).
NTHU-4 $(H_2tmdp)_2[Zn_3Ga_6O(HPO_4)(PO_4)_8]\cdot 5H_2O$	tmdp acts as a diprotonated cation.	Framework composed of corner-sharing GaO_4, MO_4 (M = Zn/Ga), and PO_4/HPO_4 polyhedra	Features a unique 3D network with 14-membered ring channels.
NTHU-8 $(Hbpy)[Zn_2PO_4(btec)(Hbpy)_2]$	btec serves as an anionic linker; Hbpy plays a dual role as a terminal ligand and template.	1D chains built of corner-sharing ZnO_3N and PO_4 tetrahedra	Features nanoscale channels and bimodal porosity.
NTHU-10 $(Hbpy)[Zn(H_2PO_4)(btec)_{0.5}]$	btec serves as an anionic ligand; Hbpy acts as a template.	0D subunits consisting of ZnO_4 and H_2PO_4 tetrahedra	The first 3D organometallophosphate structure constructed from 0D inorganic building unit
NTOU-11 (in this work) $Zn_8(HPO_4)_2(PO_4)_4(HDPBP)_2(DPBP)_3\cdot 2NO_3\cdot 2H_2O$	DPBP plays a dual role as a linker and a terminal unit	2D layers composed of 3-, 4-, and 8-membered rings.	Features a nanoscale interlayer separation exceeding 20 Å.

BDC = 1,4-benzene dicarboxylate; tmdp = 4,4’-trimethylene dipyridine; btec = benzene-1,2,4,5-tetracarboxylate; bpy = 4,4’-bipyridine; DPBP = 4,4'-di(4-pyridyl)biphenyl

Table S2. CIE chromaticity coordinates of DPBP ligand and NTOU-11 under 254 nm and 365 nm irradiation.

Wavelength (nm)	254 nm	365 nm
DPBP	(0.22, 0.16)	(0.18, 0.13)
NTOU-11	(0.22, 0.20)	(0.25, 0.28)

Table S3. The CIE coordinates of **R6G@NTOU-11** powders under 254 nm and 365 nm irradiation.

R6G concentration	0.001 mM	0.01 mM	0.05 mM	0.1 mM	0.25 mM	0.5 mM	1 mM	5 mM	10 mM	15 mM	20 mM
Under 254 nm irradiation	(0.25, 0.32)	(0.25, 0.30)	(0.27, 0.29)	(0.25, 0.28)	(0.27, 0.27)	(0.27, 0.29)	(0.26, 0.28)	(0.23, 0.23)	(0.27, 0.27)	(0.23, 0.26)	(0.30, 0.29)
Under 365 nm irradiation	(0.20, 0.22)	(0.20, 0.23)	(0.21, 0.22)	(0.21, 0.21)	(0.21, 0.21)	(0.20, 0.19)	(0.20, 0.19)	(0.20, 0.19)	(0.20, 0.18)	(0.20, 0.23)	(0.20, 0.18)