## **Supporting Information**

## Novel self-activated upconversion rare earth orthoniobate photochromics with highperformance optical storage

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**Figure S1.** XRD patterns of  $Er_xYb_{1-x}NbO_4$  solid solutions measured at (a)  $2\theta = 10-90^\circ$ , and (b)  $2\theta = 28-31^\circ$ .



**Figure S2.** XRD refinements of samples: (a)  $Er_xYb_{1-x}NbO_4$  (x=0.001), (b)  $Er_xYb_{1-x}NbO_4$  (x=0.5) b).



Figure S3. Inverse fast fourier transformation (FFT) and diffraction images of  $ErNbO_4$  sample.



Figure S4. SEM images and element mappings of  $Er_xYb_{1-x}NbO_4$  samples: (a) x=0, (b) x=0.001, (c) x=0.005, (d) x=0.01, (e) x=0.02, (f) x=0.03, (g) x=0.05, (h) x=0.1, (i) x=0.3, (j) x=0.5, (k) x=0.7, (l) x=0.9, (m) x=1. (n)-(q) Yb, Er, O, and Nb element mappings of  $Er_{0.5}Yb_{0.5}NbO_4$ , respectively.



**Figure S5.** Average grain size of Er<sub>x</sub>Yb<sub>1-x</sub>NbO<sub>4</sub> samples.



**Figure S6.** Reflection spectral changes of  $Er_xYb_{1-x}NbO_4$  samples before and after 365 nm irradiation for 10 s. The insets are surface color photographs of ceramic samples before (right) and after (left) 365 nm irradiation.



**Figure S7.** Surface color changes of coloration and decoloration process for YbNbO<sub>4</sub> sample upon 365 nm and 405 nm irradiation.



**Figure S8.** (a) UC emission spectra of  $Er_xYb_{1-x}NbO_4$  (x=0.05) sample with different pumping powers. (b) UC intensity (green and red emission) dependence on the pumping power of  $Er_xYb_{1-x}NbO_4$  (x=0.05) in a logarithmic diagram.



**Figure S9.** UC emission spectral changes of  $Er_xYb_{1-x}NbO_4$  samples before and after 365 nm irradiation.



**Figure S10.** The changes of luminescent switching contrast ( $\Delta R_t$ ) upon alternating 365 nm

Irradiation (10s) and thermal stimulus (500 °C, 10 min) under 10 cycles.



Figure S11. The changes of luminescent switching contrast ( $\Delta R_t$ ) upon alternating 365 nm

Irradiation (10s) and thermal stimulus (300 °C, 30 min) under 10 cycles.



Figure S12. (a) XPS spectra of YbNbO<sub>4</sub> sample. (b) and (c) Fitting Nb3d and Yb4d spectra before and after 365 nm irradiation.



**Figure S13.** A comparison of absorption spectrum ( $\Delta Abs$ ) (up) for YbNbO<sub>4</sub> and UC emission spectrum for Er<sub>x</sub>Yb<sub>1-x</sub>NbO<sub>4</sub> (x=0.01) (down).



Figure S14. Decay life times of Er<sub>x</sub>Yb<sub>1-x</sub>NbO<sub>4</sub> samples before and after 365 nm irradiation.



**Figure S15.** UC emission spectral changes of Tm or Ho doped YbNbO<sub>4</sub> samples before and after 365 nm irradiation: (a) Tm<sub>0.1</sub>Yb<sub>0.9</sub>NbO<sub>4</sub>, (b) Ho<sub>0.1</sub>Yb<sub>0.9</sub>NbO<sub>4</sub>.



**Figure S16.** Temperature dependence on 980 nm laser irradiation time and power density for the  $\text{Er}_{x}\text{Yb}_{1-x}\text{NbO}_{4}$  (x=0.01, 0.9, and 1),  $\text{Tm}_{y}\text{Yb}_{1-y}\text{NbO}_{4}$  (y = 0.1) and  $\text{Ho}_{z}\text{Yb}_{1-z}\text{NbO}_{4}$  (z = 0.1) samples.



Figure S17. Downshifting excitation (left) and emission (right) spectra of ErNbO<sub>4</sub> sample  $(\lambda_{ex}=378 \text{ nm}, \lambda_{em}=558 \text{ nm})$  before and after 365 nm irradiation.

Samples	λem	λex	$\Delta Rt\%$	Decoloration	Coloration	Readout	Ref.
KNN:Ho <sup>3+</sup>	453nm	551nm	77%	230°C 10min	407nm 20s	-DS	[1]
KSN:Sm <sup>3+</sup>	407nm	601nm	60%	200°C 10min	395nm 60s	-DS	[2]
BMS:Bi <sup>3+</sup>	365nm	510nm	65%	532nm 20min	254nm16min	-DS	[3]
BZO:Sm <sup>3+</sup>	406nm	596nm	90.05%	200°C 20min	254nm 8min	-DS	[4]
BiT:Pr <sup>3+</sup>	451 nm	611nm	76%	250°C 1min	405nm 5min	-DS	[5]
KNN:Pr <sup>3+</sup>	448 nm	610nm	50.71%	200°C5min	395nm 5min	-DS	[6]
SSO:Sm <sup>3+</sup>	407nm	647nm	72.2%	300°C 10min	290nm 5min	-DS	[7]
NBN:Er <sup>3+</sup> /Yb <sup>3+</sup>	980nm	557nm	85.88%	200°C 10min	407nm 10s	16%UC	[8]
NBN:Er <sup>3+</sup> /Yb <sup>3+</sup>	487nm	550nm	35%	200°C 10min	407nm (in situ)	35%DS	[8]
PWO:Er <sup>3+</sup> /Yb <sup>3+</sup>	980 nm	532nm	80%	808nm 140s	532nm 40s	-UC	[9]
SBT:Ho <sup>3+</sup> /Yb <sup>3+</sup>	980 nm	546nm	74%	200°C 1min	405nm3min	-UC	[10]
KNLNB:Er <sup>3+</sup>	980 nm	556nm	60.46%	230°C 10min	407nm 20s	-UC	[11]
NBT:Ho <sup>3+</sup> /Yb <sup>3+</sup>	980 nm	546nm	36.6%	230°C 10min	405nm 3min	-UC	[12]
NBN:Eu <sup>3+</sup>	465 nm	618nm	63%	200°C 10min	465nm (in situ)	63%DS	[13]
NBN:Pr <sup>3+</sup>	452 nm	613nm	50%	200°C 10min	452nm 50%DS (in situ)		[14]
NBN:Sm <sup>3+</sup>	406 nm	603nm	62%	200°C 10min	406nm (in situ)	62%DS	[15]
Er <sub>x</sub> Yb <sub>1-x</sub> NbO <sub>4</sub>	980nm	556nm	99.2%	405nm 5min	365nm <1s	Non	This Work

**Table S1.** Photoluminescence, luminescent modulation, response time, and decoloration and coloration

 process in some representative inorganic photochromic materials.

Readout: destruction degree with DS or UC readout mode. -: No report; Non: non-destructive readout.

Lattice O(O <sup>2-</sup> )	Absorb (Vo)	Vo/ O <sup>2-</sup>
523.29 eV	531.16 eV	0.189
529.19 eV	530.951 eV	0.352
	529.19 eV	529.19 eV 530.951 eV

Table S2. Fitting parameters of O1s-XPS spectra of YbNbO4 before and after irradiation at 365 nm

\*Vo means O vacancy

Table S3. Fitting parameters of the decay lifetime curves of  $Er_xYb_{1-x}NbO_4$  samples.

Samples	$\tau_1(\mu s)$		$\tau_2(\mu s)$		χ2		$ au_A(\mu s)$	
	before	after	before	after	before	after	before	after
x=0.0001	30.6	6.1	65.1	24.3	1.023	1.354	43.4	8.5
x=0.005	25.8	5.1	46.7	12.6	1.094	1.009	34.1	5.9
x=0.01	26.5	5.4	46.1	14.1	0.940	0.990	36.1	6.2
x=0.02	28.1	5.2	50.1	15.9	0.999	1.057	39.5	5.9
x=0.03	26.3	5.4	46.1	11.3	1.036	1.037	36.8	6.2
x=0.05	19.6	5.7	38.4	8.1	1.021	0.841	30.1	7.2
x=0.1	19.0	7.2	39.6	11.1	0.934	0.909	29.7	7.7
x=0.3	14.7	6.7	32.5	9.6	1.080	1.053	23.1	7.6
x=0.5	15.8	6.8	50.2	10.7	0.689	0.916	34.6	8.9

x=0.7	14.7	8.5	48.8	14.4	0.762	0.868	31.7	10.3
x=0.9	42.5	11.7	83.7	20.3	0.984	0.894	66.9	14.1
x=1	42.3	15.7	80.6	26.8	0.959	0.836	66.1	19.7

## References

1 Y. Zhang, J. Liu, H. Q. Sun, D. F. Peng, R. H. Li, C. K. Bulin, X. S. Wang, Q. W. Zhang and X. H. Hao, J. Am. Ceram. Soc., 2018, 101(6), 2305.

2 S. Y. Cao, Q. Chen, J. T. Liu, C. Y. Wu, L. L. Li, J. Xu, G. H. Cheng and F. Gao, , J. Eur. Ceram. Soc., 2020, 40(15), 6061.

3 Y. T. Ren, Z. W. Yang, Y. H. Wang, M. J. Li, J. B. Qiu, Z. G. Song, J. Yu, A. Ullah and I. Khan, Sci. China Mater., 2020, 63(4), 582.

4 R. T. Zhang, Y. H. Jin, L. F. Yuan, H. Y. Wu, G. T. Xiong, C. L. Wang, L. Chen and Y. H. Hu, J. Lumin., 2021, 233, 117922.

5 T. Wei, F. M. Yang, B. Jia, C. Z. Zhao, L. Y. Liu, H. J. Zhang, Y. Zhang, J. F. Zhang, X. T. Yan and J. T. Yang, Ceram. Int., 2020, 46(11), 18507.

6 Q. Zhang, L. H. Luo, J. Gong, P. Du, W. P. Li and G. L. Yuan, J. Eur. Ceram. Soc., 2020, 40(12), 3946.

7 J. Tang, P. Du, W. P. Li, G. L. Yuan, Z. F. Liu and L. H. Luo, Chem. Eng. J., 2021, 410, 128287.

8 Q. W. Zhang, S. S. Yue, H. Q. Sun, X. S. Wang, X. H. Hao and S. L. An, J. Mater. Chem. C, 2017, 5(15), 3838.

9 X. Bai, Z. W. Yang, Y. H. Zhan, Z. Hu, Y. T. Ren, M. J. Li, Z. Xu, A. Ullah, I. Khan, J. B. Qiu, Z. G. Song, B. T. Liu and Y. H. Wang, ACS Appl. Mater. Interfaces, 2020, 12(19), 21936.

10 J. Q. Bi, T. Wei, L. H. Shen, F. M. Yang, C. Z. Zhao, M. C. Wang, Q. Q. Yang and Y. X. Lin, J. Am. Ceram. Soc., 2021, 104(4), 1785.

11 H. Q. Sun, Y. Lv, Y. Zhu, J. F. Lin, X. Wu, Q. W. Zhang and X. H. Hao, J. Am. Ceram. Soc., 2019, 102(11), 6732.

12 Z. H. Lu, K. X. Li, J. Wang, L. H. Luo, Opt. Mater., 2021, 111, 110718.

13 Q. W. Zhang, H. Q. Sun, H. Li, X. S. Wang, X. H. Hao, J. L. Song, S. L. An, Chem. Commun., 2015, 51, 16316.

14 Q. W. Zhang, H. Q. Sun, X. S. Wang, X. H. Hao and S. L. An, ACS Appl. Mater. Interfaces, 2015, 7, 25289.

15 Q. W. Zhang, Y. Zhang, H. Q. Sun, Q. Sun, X. S. Wang, X. H. Hao and S. L. An, J. Eur. Ceram. Soc., 2017, 37, 955.