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## **Supporting Information**

## First observation of mutual energy transfer of Mn<sup>4+</sup>-Er<sup>3+</sup> via different excitation in Gd<sub>2</sub>ZnTiO<sub>6</sub>:Mn<sup>4+</sup>/Er<sup>3+</sup> phosphors

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**Fig. S1** NIR emission spectra of (a)  $Gd_{2(1-x)}ZnTi_{0.998}O_6:Mn_{0.002}$ ,  $Er_{2x}$  (x =0, 0.005, 0.01, 0.03, 0.05, 0.07, 0.09) and (b)  $Gd_{1.98}ZnTi_{1-y}O_6:Mn_y$ ,  $Er_{0.02}$  (y =0, 0.0005, 0.001, 0.002, 0.004, 0.006, 0.008, 0.01, 0.015, 0.02) excited at 335 nm light. The inset shows the  $Er^{3+}$  and  $Mn^{4+}$ concentration dependent 1529 nm intensity of  $Er^{3+}$ .



Fig. S2 Excitation and emission spectra of the Mn<sup>4+</sup> single-doped GZT sample.



**Fig. S3** The typical Mn<sup>4+</sup> and Er<sup>3+</sup> emission spectra of GZT: $0.002Mn^{4+}/0.02Er^{3+}$  under common excitation wavelength 379 nm (corresponding to the  ${}^{4}A_{2} \rightarrow {}^{4}T_{1}$  of Mn<sup>4+</sup> and  ${}^{4}I_{15/2} \rightarrow {}^{4}G_{11/2}$  of Er<sup>3+</sup>).



Fig. S4 Raman spectra of the GZT matrix



**Fig. S5** Decay curves of Mn<sup>4+</sup> luminescence in GZT: 0.002Mn<sup>4+</sup>, 0.02Er<sup>3+</sup> (y =0.0005, 0.001, 0.002, 0.004, 0.006, 0.008, 0.01, 0.015, 0.02) monitored at 705 nm excited at 335 nm light



Fig. S6 Emission spectra of  $Mn^{4+}$  in  $Gd_{2(1-x)}ZnTi_{0.998}O_6$ :  $_{0.002}Mn^{4+}$ ,  $2xEr^{3+}$  (x =0, 0.005, 0.01, 0.03, 0.05, 0.07, 0.09) excited at 335 nm light.

Er (x)	A <sub>1</sub>	$\tau_1^{}$ (ms)	A <sub>2</sub>	τ <sub>2</sub> (ms)	τ <sub>av</sub> (ms)	$\eta_{_{ m ET}}$
0	21928.10 1	0.701	/	/	0.701	0
0.05	35732.45 8	0.677	2737.676	0.07	0.672	0.041
0.01	28787.43 4	0.623	6723.674	0.147	0.598	0.147
0.03	18249.24 4	0.496	16306.33 6	0.131	0.426	0.392
0.05	8737.664	0.388	17427.25 6	0.101	0.290	0.586
0.07	6241.655	0.288	16821.62 8	0.072	0.201	0.713
0.09	2895.307	0.216	11685.86 4	0.054	0.135	0.807

**Table S1** Average lifetime ( $\tau$ ) of Mn<sup>4+</sup> 705 nm emission and the energy transfer efficiencies ( $\eta_{ET}$ ) from Mn<sup>4+</sup> to Er<sup>3+</sup> in Gd<sub>2(1-x)</sub>ZnTi<sub>0.998</sub>O<sub>6</sub>:0.002Mn<sup>4+</sup>, 2xEr<sup>3+</sup> phosphors

Mn(y)	$A_1$	$ au_1$	A <sub>2</sub>	τ2	τ <sub>av</sub> (ms)
0.005	34912.354	0.637	5391.481	0.151	0.620
0.001	32495.059	0.623	5347.319	0.079	0.612
0.002	28787.434	0.623	6723.674	0.147	0.598
0.004	29396.155	0.596	7338.239	0.136	0.571
0.006	23168.134	0.554	8817.416	0.132	0.519
0.008	20022.614	0.532	8777.169	0.122	0.495
0.01	12893.303	0.465	7253.600	0.104	0.425
0.015	9703.535	0.436	6904.3966	0.090	0.392
0.02	3406.119	0.285	3804.534	0.051	0.246

**Table S2** Average lifetime ( $\tau$ ) of Mn<sup>4+</sup> 705 nm emission in Gd<sub>1.98</sub>ZnTi<sub>1-y</sub>O<sub>6</sub>: yMn<sup>4+</sup>,0.02Er<sup>3+</sup>phosphors (under 335 nm excitation)

**Table S3** Average lifetime ( $\tau$ ) of Er<sup>3+</sup> 551 nm emission and the energy transfer efficiencies ( $\eta_{\text{ET}}$ ) from Er<sup>3+</sup> to Mn<sup>4+</sup> in Gd<sub>1.98</sub>ZnTi<sub>1-y</sub>O<sub>6</sub>:yMn<sup>4+</sup>, 0.02Er<sup>3+</sup> phosphors (under 980 nm pulse laser excitation)

Mn (y)	A <sub>1</sub>	τ <sub>1</sub> (ms)	A <sub>2</sub>	τ <sub>2</sub> (ms)	τ <sub>av</sub> (ms)	$\eta_{_{ m ET}}$
0	2131.085	0.383	5571.333	0.080	0.276	0
0.0005	2016.008	0.326	5662.927	0.068	0.231	0.163
0.001	1978.692	0.293	5598.314	0.059	0.208	0.246
0.002	2103.444	0.208	5546.871	0.040	0.152	0.449
0.004	2083.221	0.197	5384.386	0.037	0.145	0.475
0.006	2232.884	0.134	5536.619	0.024	0.100	0.638
0.008	2115.080	0.125	5584.994	0.021	0.093	0.663
0.010	1910.934	0.091	5640.007	0.016	0.065	0.764
0.015	1909.984	0.076	5596.935	0.013	0.055	0.801
0.020	2083.261	0.018	5879.827	0.001	0.016	0.942