## **Supporting Information**

## Photo-Induced Magnetic Hysteresis in a Cyanide-bridged Two-dimensional [Mn<sub>2</sub>W] Coordination Polymer

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T/K	120 K		
Formula	$C_{32}H_{44}Mn_2N_{12}O_4S_4W$		
CCDC	2130620		
Fw	1170.68		
Crystal system	Monoclinic		
Space group	<i>C</i> 2/c		
<i>a</i> (Å)	22.1413(7)		
<i>b</i> (Å)	19.9940(8)		
<i>c</i> (Å)	10.4520(3)		
α (°)	90		
$eta(^{ m o})$	109.911(1)		
γ(°)	90		
$V(Å^3)$	V(Å <sup>3</sup> ) 4350.4(3)		
Ζ	4		
$ ho_{ m calc}$ (g/cm <sup>3</sup> )	1.769		
Reflections collected	16555		
Unique reflections $(R_{int})$	0.0504		
Goodness-of-fit on $F^2$	1.021		
$R_1, [I \ge 2\sigma(I)]$	0.0382		
$wR_2$ , $[I \ge 2\sigma(I)]$	0.0841		

Table S1. Crystallographic data and Structure Refinements for compound  $[Mn_2W_{LS}]$ .

 $R_{1} = \Sigma (|F_{o}| - |F_{c}|) / \Sigma |F_{o}|; wR_{2} = [\Sigma w (|F_{o}| - |F_{c}|)^{2} / \Sigma w F_{o}^{2}]^{1/2}$ 

		S <sub>DD</sub>	S <sub>SAPR</sub>	S <sub>BTPR</sub>
[Mn <sub>2</sub> W <sub>LS</sub> ]		1.763, 2.856	0.892, 1.062	2.314, 2.739
<b>Mn<sub>2</sub>W</b> (ref. 57)		2.241; 2.496	0.113; 0.118	2.029; 2.186
(H <sub>4</sub> cyclam)[W(CN) <sub>8</sub> ] ·1.5H <sub>2</sub> O (ref. 61)		1.023	0.903	1.318
<b>Zn<sub>6</sub>W<sub>3</sub></b> (ref. 46)	W1	1.110	0.969	1.151
	W2	1.922	0.295	1.540
	W3	1.781	0.532	1.355
			S <sub>OC</sub>	$S_{TPR}$
$[Mn_2W_{LS}]$	Mn1		0.131	16.536
	Mn1		0.427	14.368

Table S2. SHAPE calculations results.

 ${}^{a}S_{DD}$ ,  $S_{SAPR}$ ,  $S_{BTPR}$ ,  $S_{OC}$ ,  $S_{TPR}$ -shape measure relatives to the triangular dodecahedron (DD), square antiprism (SAPR), biaugmented trigonal prism (BTPR), octahedron (OC) and trigonal prism (TPR). When the respective shape measure parameter equals zero, the real geometry coincides with the idealized one.



Figure S1. Powder X-ray diffraction (PXRD) pattern (olive) of  $[Mn_2W_{LS}]$  obtained at 298 K in comparison to the pattern (blue) of  $[Mn_2W_{LS}]$  calculated from the crystal structures determined at 120 K.



Figure S2. TGA result of compound  $[Mn_2W_{LS}]$  in nitrogen atmosphere at 10 K min<sup>-1</sup>. The 8% of weight lost is in agreement with the losing two water molecules and two methanol molecules in compound  $[Mn_2W_{LS}]$ .

## Magnetic measurement for [Mn<sub>2</sub>W].

First, the freshly prepared [ $Mn_2W_{LS}$ ] compound was quenched in MPMS at 150 K without purging the chamber. Then cooling down to 2 K and the susceptibility data were collected in the sequence of 2 K $\rightarrow$ 300 K $\rightarrow$ 2 K (2 K min<sup>-1</sup>). Then the isothermal magnetization curve was measured at 3 K.

Next, the photomagnetic measurement on  $[Mn_2W_{LS}]$  (0.9 mg) was performed at 10 K in a DC field of 1000 Oe under 405-nm diode laser light. We increased the laser output power when the irradiation duration reached 12 h in anticipation of boosting the speed of irradiation response. After 18 hours of irradiation, hysteresis loops were measured at 2, 10, and 15 K at a sweeping rate of 25 Oe s<sup>-1</sup> with the laser light turned off. Subsequently, the ZFC-FC magnetic susceptibility was measured on heating in the range of 2–30 K in a 100 Oe DC magnetic field after zero-field cooling from 20 to 2 K at a sweeping rate of 2 K min<sup>-1</sup>. The temperature dependence of AC susceptibility was then measured in the range from 2 to 35 K under a 0 Oe DC magnetic field and a 3.5 Oe AC magnetic field, respectively.

Finally, the temperature dependence of DC magnetic susceptibility of  $[Mn_2W_{HS}]$  was measured in the sequence of 2 K $\rightarrow$ 300 K $\rightarrow$ 2 K (at a sweeping rate of 2 K min<sup>-1</sup>) under 1000 Oe. Following that, the isothermal magnetization curve was recorded at 2 K.



Figure S3. Temperature dependence of the  $\chi T$  values for  $[Mn_2W_{LS}]$  before irradiation under a DC field of 1000 Oe in heating and cooling modes.



Figure S4. Magnetization vs magnetic field curve for  $[Mn_2W_{LS}]$  at T = 3.0 K measured before irradiation.



**Figure S5.**  $\chi T vs$  time dependence recorded for [**Mn**<sub>2</sub>**W**] at T = 10 K and H = 0.1 T during the irradiation experiment using 405 nm light.



Figure S6. Magnetization *vs* magnetic field curve for  $[Mn_2W]$  at T = 2.0 K measured after and before 405 nm irradiation.



**Figure S7.** Temperature dependence of the  $\chi T$  values for [**Mn**<sub>2</sub>**W**], before irradiation (blue), after irradiation (orange) and after heating (olive) to 300 K under a DC field of 1000 Oe. Inset: enlargement view of low-temperature region of the  $\chi T$  vs. *T* plots.



Figure S8. Temperature dependence of the in-phase ( $\chi'$ ) and out-of-phase ( $\chi''$ ) of AC magnetic susceptibilities for [Mn<sub>2</sub>W<sub>HS</sub>] after 405 nm light irradiation under zero DC field and 3.5 Oe AC field.