## <Supporting Information>

## Magnetoluminescent bifunctional dysprosium-based phosphotungstates

## with synthesis and correlations between structures and properties

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## **Electronic Supporting Information**

Figure S1. Ball-and-stick representation of the polyanions for 1Dy (a), 2Dy (b), 3Dy(c) and 4Dy.

- Figure S2. The excitation spectra of solid state **1Dy–4Dy** in the visible (a–d) or near-infrared (e–h) region at room temperature.
- Figure S3. The emission spectra of 2Dy–4Dy in the visible (a) or near-infrared (b) region under the same conditions.
- Figure S4. IR spectra of the 1Dy and 2Dy compared with TMA counter cations.
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- Figure S9. The thermal behaviors of the 3Dy(a) and 4Dy(b) between 20 and 800 °C.
- **Figure S10.** Temperature dependency of the in-phase ( $\chi_M$ ') and out-of-phase ( $\chi_M$ '') signals from the AC susceptibility measurements of **1Dy**.
- **Figure S11.** Temperature dependency of the in-phase ( $\chi_M$ ') and out-of-phase ( $\chi_M$ '') signals from the AC susceptibility measurements of **2Dy**.
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- Figure S14. Hysteresis loops for 1Dy (a) and 2Dy (b) measured at different temperatures with sweeping rate of 500 Oe/s.
- Figure S15. Hysteresis loops for 1Dy (a–c), 2Dy (d–f), 3Dy (g–i) and 4Dy (h–l) measured at different sweeping rates (500, 300 and 100 Oe/s) at 2 K.
- Figure S16. EDX spectra of single crystals of 1Dy-4Dy.



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**Figure S2.** The excitation spectra of solid state **1Dy–4Dy** in the visible (a–d) or near-infrared (e–h) region at room temperature.



**Figure S3.** The emission spectra of **2Dy–4Dy** in the visible (a) or near-infrared (b) region under the same conditions.



**Figure S4.** IR spectra of the **1Dy** and **2Dy** compared with TMA counter cations, which display the typical vibration bands of monolacunary Keggin-type polyanion  $[\alpha$ -PW<sub>11</sub>O<sub>39</sub>]<sup>7-</sup> and organic components in products.



**Figure S5.** IR spectra of the **3Dy** compared with Na<sub>2</sub>CO<sub>3</sub> cations, which display the typical vibration bands of trivacant Keggin-type polyanion  $[\alpha$ -PW<sub>9</sub>O<sub>34</sub>]<sup>9-</sup> and carbonyl.



**Figure S6.** IR spectrum of the **4Dy** displays the typical vibration bands of divacant Keggin-type polyanion  $[\alpha$ -PW<sub>10</sub>O<sub>38</sub>]<sup>11-</sup>.



**Figure S7.** X-ray powder diffraction patterns of **1Dy–4Dy** revealing the good coincidence between experiment values and stimulated values.



**Figure S8.** The thermal behaviors of the **1Dy**(a) and **2Dy**(b) have been investigated between 20 and 1000 °C. Their TG curves show three mass loss stages.



Figure S9. The thermal behaviors of the 3Dy(a) and 4Dy(b) have been investigated between 20 and 800 °C.



**Figure S10.** Temperature dependency of the in-phase  $(\chi_M')$  and out-of-phase  $(\chi_M'')$  signals from the AC susceptibility measurements of **1Dy** with an applied static field of 0 Oe (a) and (b), 1000 Oe (c) and (d), 2000 Oe (e) and (f), 3000 Oe (g) and (h).



**Figure S11.** Temperature dependency of the in-phase  $(\chi_M')$  and out-of-phase  $(\chi_M'')$  signals from the AC susceptibility measurements of **2Dy** with an applied static field of 0 Oe (a) and (b), 1000 Oe (c) and (d), 2000 Oe (e) and (f), 3000 Oe (g) and (h).



**Figure S12.** Temperature dependency of the in-phase  $(\chi_M')$  and out-of-phase  $(\chi_M'')$  signals from the AC susceptibility measurements of **3Dy** with an applied static field of 0 Oe (a) and (b), 1000 Oe (c) and (d), 2000 Oe (e) and (f), 3000 Oe (g) and (h).



**Figure S13.** Temperature dependency of the in-phase  $(\chi_M')$  and out-of-phase  $(\chi_M'')$  signals from the AC susceptibility measurements of **4Dy** with an applied static field of 0 Oe (a) and (b), 1000 Oe (c) and (d), 2000 Oe (e) and (f), 3000 Oe (g) and (h).



Figure S14. Hysteresis loops for 1Dy (a) and 2Dy (b) measured at different temperatures with sweeping rate of 500 Oe/s.



**Figure S15.** Hysteresis loops for **1Dy** (a–c), **2Dy** (d–f), **3Dy** (g–i) and **4Dy** (h–l) measured at different sweeping rates (500, 300 and 100 Oe/s) at 2 K



Figure S16. EDX spectra of single crystals of 1Dy-4Dy.

We used EDX-SEM to characterize the composition of these compounds. In this regard, these compounds were identified using single crystals. Figure S16 show the EDX spectra recorded for **1Dy–4Dy**, respectively, which evidently indicate the presence of P, W and Dy main components.