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## Three novel mononuclear Mn(III) based magnetic materials with square pyramidal versus octahedral

## geometries<sup>+</sup>

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**Fig. S1**: The H-bonding direction and CH $\cdots\pi$  interactions observed in Complex **1** (above). The molecular view of 2D network through H-bonding and 3D framework through H-bonding interactions in complex **1** (below).



Fig. S2: The H-bonding direction and CH… $\pi$  interactions observed in complex 2.





**Fig. S3**: The molecular view of H-bonding (orange dot line),  $CH\cdots\pi$  (green dot line) and  $\pi\cdots\pi$  (yellow dot line) interactions in **3** (above). The 3D framework through H-bonding and  $CH\cdots\pi$  interactions in complex **3** (below).



Fig. S4: IR spectrum of complex 1.



Fig. S6: IR spectrum of complex 3.

The experimental magnetic susceptibility and magnetization data of **1**, **2** and **3** were simultaneously analyzed by using the appropriate spin Hamiltonian for a dinuclear model [Eq. (1)]

$$\hat{H} = -2J\hat{S}_{1}\hat{S}_{2} + D(\hat{S}_{1,z}^{2} + \hat{S}_{2,z}^{2}) + \mu_{B}g_{iso}H(\hat{S}_{1} + \hat{S}_{2})$$
(1)



**Fig. S7**: Temperature dependence of  $\chi T$  for **1** ( $\bigcirc$ ) under an applied field of 1000 Oe (up left), the solid line is the best-fit curve (see text). Field dependence of the magnetization as M vs H/T (up right) and M vs H (down) plots for **1** between 2 and 5 K, the solid lines are the best-fit curves (up right).





**Fig. S8**: Temperature dependence of  $\chi T$  for **2** ( $\bigcirc$ ) under an applied field of 1000 Oe (up left), the solid line is the best-fit curve (see text). Field dependence of the magnetization as M vs H/T (up right) and M vs H (down) plots for **2** between 2 and 5 K, the solid lines are the best-fit curves (up right).



**Fig. S9**: Temperature dependence of  $\chi T$  for **3** ( $\bigcirc$ ) under an applied field of 1000 Oe (up left), the solid line is the best-fit curve (see text). Field dependence of the magnetization as M vs H/T (up right) and M vs H (down) plots for **2** between 2 and 5 K, the solid lines are the best-fit curves (up right).











**Fig. S12.** Frequency dependence of the real ( $\chi'$ , left) and imaginary ( $\chi''$ , right) parts of the *ac* susceptibility for a polycrystalline sample of **2** at 1.8 K at different *dc*-field between 0 and 1000 Oe. Solid lines are eye guides.



**Fig. S13.** Temperature (left) and frequency (right) dependence of the real ( $\chi'$ , top) and imaginary ( $\chi''$ , bottom) components of the *ac* susceptibility at different *ac* frequencies between 1 and 1500 Hz and different temperatures between 1.8 and 8 K, respectively, with a 3 Oe *ac* field for a polycrystalline sample of **3** in a zero *dc*-field.



**Fig. S14.** Frequency dependence of the real ( $\chi'$ , left) and imaginary ( $\chi''$ , right) parts of the *ac* susceptibility for a polycrystalline sample of **3** at 1.8 K at different *dc*-field between 0 and 3000 Oe. Solid lines are eye guides.