Observation of divergent isotope effects as well as metal ion-modulated T_C and spin-canting nature in isostructural supramolecular magnets

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Figure S1 FT-IR spectra of (a) **1** and **2** (b) **3** and **4**. The spectroscopy changes owing to the deuteration in the region of 2320-2585 cm⁻¹ showed inset.

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Figure S2 ¹H NMR spectra for [4-NH₂-PyD]₂[Pt(mnt)₂].



Figure S3 Packing diagram of **1** projected along *c*-axis, which shows the separated layered arrangements of both anions and cations



Figure S4 Packing diagrams of **2** projected along crystallographic *b*- and *c*-axes, respectively as well as hydrogen bond interactions between anions and cations (#1 = x, 1+y, z; #2 = -x, 1+y, 1.5-z; #3 = x, 2-y, 0.5+z; #4 = x, -1+y, z; #5 = -x. -y, 1-z).



Figure S5 Packing diagrams of 4 projected along crystallographic *b*- and *c*-axes, respectively as well as hydrogen bond interactions between anions and cations (#1 = x, y, -1+z; #2 = 0.5-x, 0.5-y, -z; #3 = x, 1+y, -1+z; #4 = 0.5-x, 1.5-y, -z; #5 = -x, -1+y, 0.5-z; #6 = x, 1+y, z).



Figure S6 Three peaks with a Gaussian line shape were fitted using the fit-multi-peaks technique for the real part of the ac magnetic susceptibility of **2**, and the best fit gave three peak temperatures being 11.9, 13.6 and 14.5 K, respectively.





Figure S7 Powder X-ray diffraction patterns for **1-4** (red lines: experimental data; bluelines: simulation).



Figure S8 Comparison of powder X-ray diffraction patterns between 1 and 2 as well as between 3 and 4.



Figure S9 Spin density distributions of the Pt(2)-type $[Pt(mnt)_2]^-$ in **1** and Ni(2)-type $[Ni(mnt)_2]^-$ in **4**.