Supplementary Information

Intercalation of Two-Dimensional Oxalate-Bridged Molecule-Based Magnets into Layered Double Hydroxide Hosts

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- 300 nm MAG: 90000 x (b) 200 nm MAG: 110000 x
- SI 1. SEM images showing the particle's size and morphology of the ZnAl-CO₃ LDH (1, a) and the nitrate-intercalated LDH (2, b).

SI 2. Metal-to-metal ratio of the pristine ZnAl-CO₃ LDH (1), the nitrate-intercalated LDH (2) and the hybrid material (3), as estimated from EDAX analysis.

| | Al/Zn | Al/Cr | Cr/Mn |
|---|-------|-------|-------|
| 1 | 0.52 | - | - |
| 2 | 0.54 | - | - |
| 3 | 0.51 | 2.92 | 1.54 |



SI 3. *Top*: FT-IR spectra of **2** (black) and **3** (blue). *Bottom*: zoom-in of the 2000-500 cm⁻¹ region.

| Vibration mod | es [cm ⁻¹ | ¹] 2 | 3 |
|---------------|----------------------|------------------|---------------|
| υ[O-H] | | 3530, 3430 | 3457 |
| υ[C-O] | as | - | 1681, 1616 |
| | S | - | 1324, 1261 |
| v[N-O] | | 1379, 1750 | 1390 |
| v[M-O] | | 618 | 669 |
| δ[O-M-O] | | 551, 426 5 | 545, 482, 427 |

SI 4. Assignment of the main vibration modes for 2-3.



SI 5. TG/DTA curves of **2** (*top*) and **3** (*bottom*).

SI 6. Water content as derived from the thermogravimetric analysis and estimated molecular formula of the LDH-NO₃ precursor (**2**) and the restacked material (**3**).

| Water content | | | |
|---------------|------------|-------|--|
| I | Vt. loss 9 | %Mol. | Molecular formula |
| 2 | 6.32 | 1.2 | $[Zn_{1.95}Al_{1.05}(OH)_6](NO_3)_{1.05} \cdot 1.2H_2O$ |
| 3 | 8.90 | 2.5 | $[Zn_{2.12}Al_{1.08}(OH)_6][Mn_{0.24}{Cr(ox)_3}_{0.37}](NO_3)_{0.85}\bullet 2.5H_2O$ |



SI 7. Best fitting (red line) of the experimental data to a Curie-Weiss law in the 150-300 K interval.



SI 8. Field dependence of the magnetization of **3** at 2K.



SI 9. Top: Hysteresis loop of **3** measured at 2 K. Bottom: zoom-in showing the low field area. Solid lines are only a guide to the eye.

SI 10. UV-Vis spectra of the emulsions resulting from the exfoliation of ZnAl-LDH at different concentrations: 1 (green), 0.86 (orange), 0.73 (lavanda), 0.63 (pink), 0.55 (blue), 0.47 (grey), 0.41 g.dm⁻³ (black). The inset shows the linear relationship between the absorbance at 278 nm and the LDH content.

