Supporting Information for

## Assembly of alternating spin-chains with magnetically anisotropic cobalt(II) dimers

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Fig. S1. Intermolecular hydrogen-bonding interactions in 1.



Fig. S2. Interchain hydrogen-bonding interactions and potential  $\pi$ - $\pi$  stacking interactions in 2.



Fig. S3. Interchain  $\pi$ - $\pi$  stacking and hydrogen-bonding interactions in 3.



Fig. S4. Isothermal magnetization of 2 measured at 2 K.



Fig. S5. Isothermal magnetization of 3 measured at 2 K. Inset: temperature-dependent susceptibility data measured at indicated external dc fields.



**Fig. S6.** Zero-field cooled (ZFC) and field-cooled (FC) magnetization of **3** measured under an external field of 100 Oe.

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