## **Supporting Information**

## **Regulating Thermal Expansion of [FePt(CN)<sub>4</sub>] Layer by Axial Coordination and Dimensional Reduction**

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Figure S1. Thermogravimetric Differential Scanning Calorimetry (TG-DSC) curve for FePt\_py.



Figure S2. Thermogravimetric Differential Scanning Calorimetry (TG-DSC) curve for FePt\_pyz.



Figure S3. Thermogravimetric curve for FePt\_pyz·I.



**Figure S4.** Rietveld Refinement to the synchrotron powder X-ray diffraction pattern of FePt\_py at (a) 104 K and (c) 225 K in LS state and (b) 255 K and (d) 475 K in HS state.



**Figure S5.** Rietveld Refinement to the synchrotron powder X-ray diffraction pattern of FePt\_pyz at (a) 104 K and (c) 250 K in LS state and (b) 315 K and (d) 475 K in HS state.



**Figure S6.** Rietveld Refinement to the synchrotron powder X-ray diffraction pattern of FePt\_pyz·I, at (a) 104 K and (c) 380 K in LS state and (b) 410 K and (d) 475 K in HS state.



**Figure S7.** Temperature dependence of cell parameters *c* and V of FePt\_py (a, d), FePt\_pyz (b, e), and FePt\_pyz•I (c, f)



Figure S8. Variable temperature Raman spectra of FePt\_pyz.



Figure S9. Variable temperature Raman spectra of FePt\_pyz·I.