**Supporting Information Dalton Transactions**

**A rare example of double metamagnetic transition leading to 2D and 3D long-range order in the two-dimensional pyrazine- and azido-bridged cobalt(II) compound [Co(pyz)(N₃)₂]**

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**Comments X-ray powder Diffraction**

**X-ray powder diffraction**

The crystalline sample was checked for phase purity with XRPD. This measurement confirm that the sample correspond to the simulated from the single crystal X-ray structure (**Figure S1**).
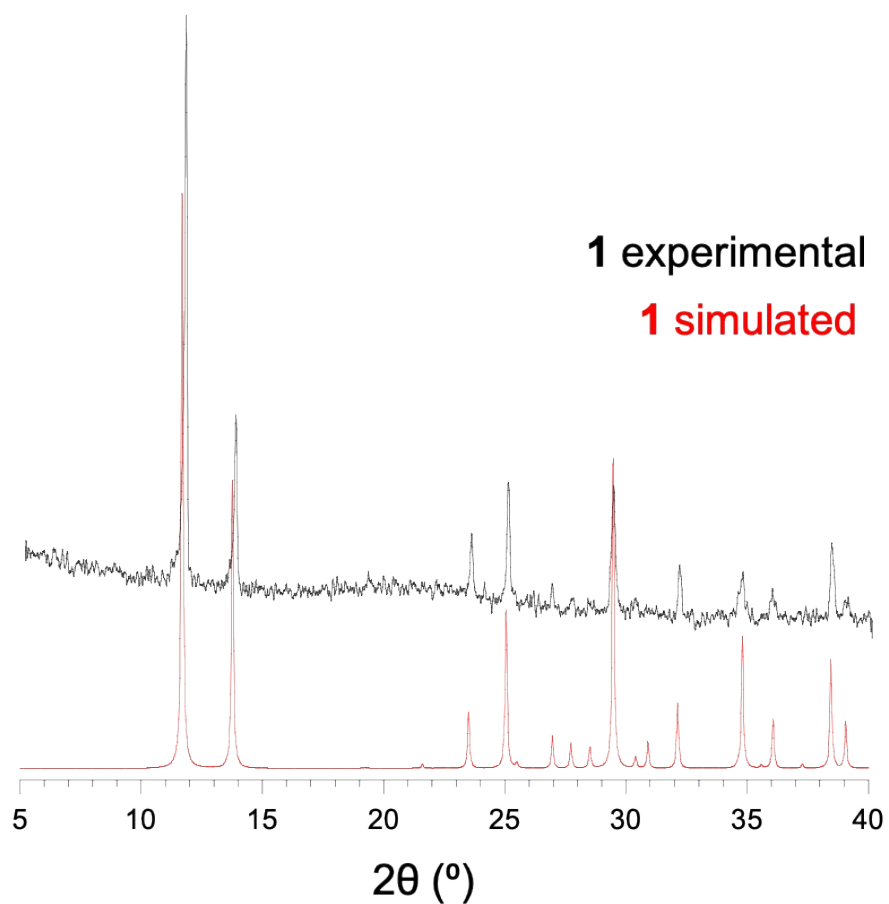


**Figure S1**. Experimental and simulated X-ray powder diffractogram for **1**.
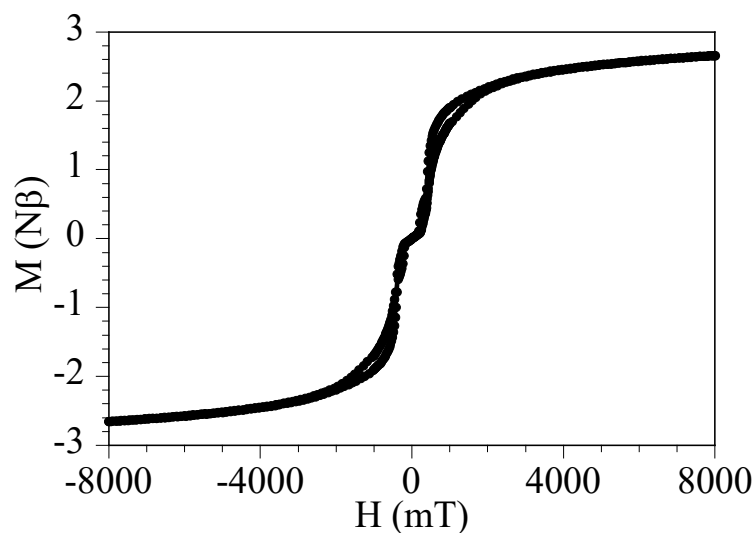
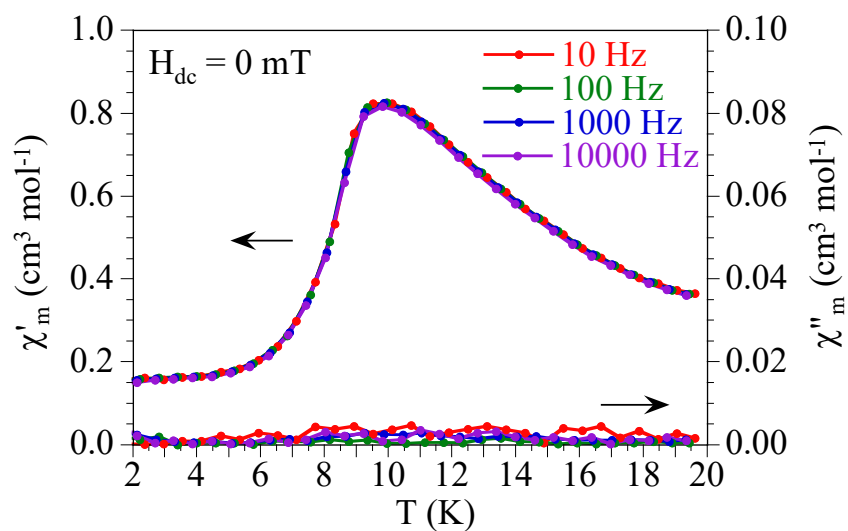**Figure S2**. Hysteresis cycle of the isothermal magnetization at 2 K for **1**.



**Figure S3**. Thermal variation of the in phase ($\chi'_m$, left scale) and out-of-phase ($\chi''_m$, right scale) susceptibilities of **1** at different frequencies with no DC field applied.

**Figure S4**. Thermal variation of the in phase ($\chi'_m$, left scale) and out-of-phase ($\chi''_m$, right scale) susceptibilities of **1** at different frequencies with a DC field of 300 mT.



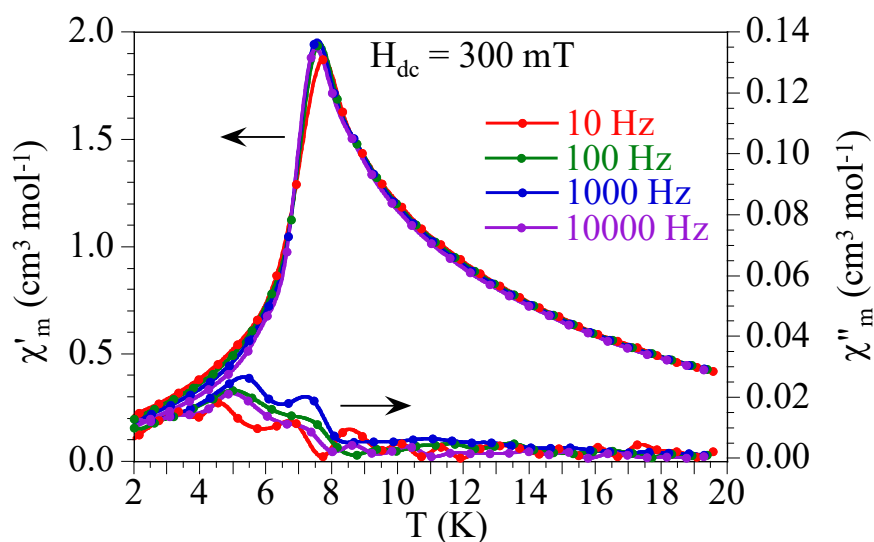**Figure S5**. Thermal variation of the in phase ($\chi'_m$, left scale) and out-of-phase ($\chi''_m$, right scale) susceptibilities of **1** at different frequencies with a DC field of 350 mT.
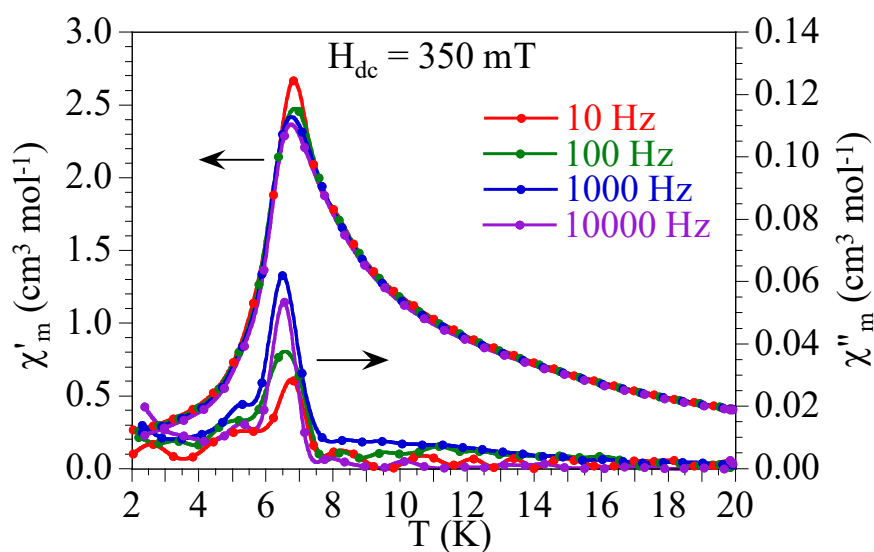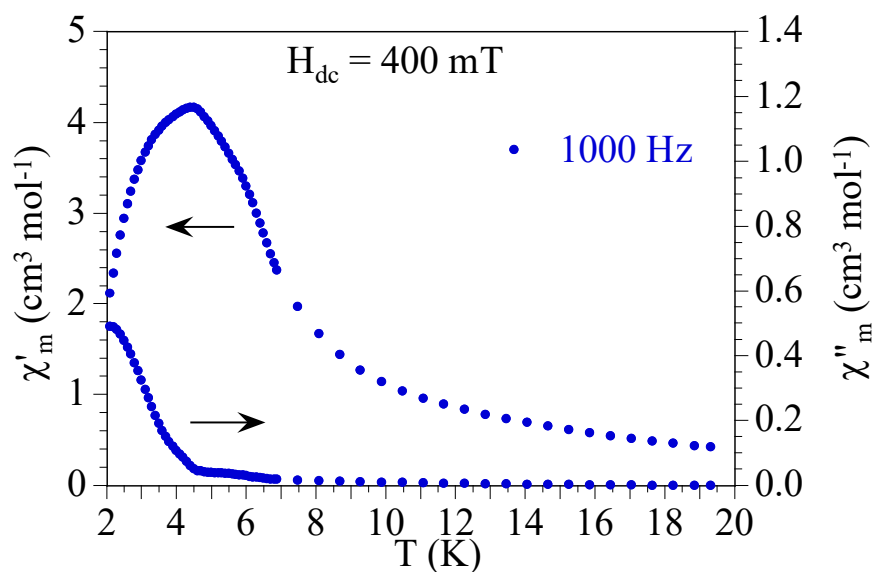
**Figure S6**. Thermal variation of the in phase ($\chi'_m$, left scale) and out-of-phase ($\chi''_m$, right scale) susceptibilities of **1** at 1000 Hz with a DC field of 400 mT.
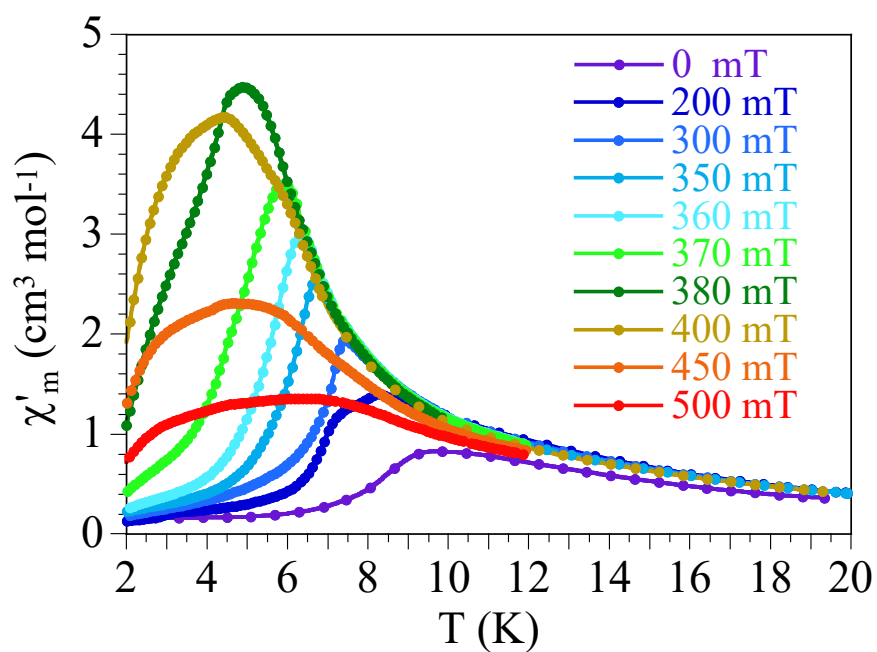


**Figure S7**. Thermal variation of the in-phase susceptibility ($\chi''_m$) for **1** at 1000 Hz with different DC fields.