

Electronic Supplementary Information:

Solid-State Transformation of $[\text{Co}(\text{NCS})_2(\text{pyridine})_4]$ into $[\text{Co}(\text{NCS})_2(\text{pyridine})_2]_n$: From Curie-Weiss Paramagnetism to Single Chain Magnetic Behaviour

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- ❖ IR spectra of compound $[\text{Co}(\text{NCS})_2(\text{pyridine})_4]$ (**1**) and $[\text{Co}(\text{SCN})_2(\text{pyridine})_2]_n$ (**2**).
- ❖ Experimental and calculated X-ray powder pattern for compound $[\text{Co}(\text{SCN})_2(\text{pyridine})_4]$ (**1**).
- ❖ Results of the magnetic measurements for compound $[\text{Co}(\text{NCS})_2(\text{pyridine})_2]_n$ (**2**) by plots of in-phase χ_M' (black) and out-of-phase χ_M'' (red) ac magnetic susceptibility as function of frequency.

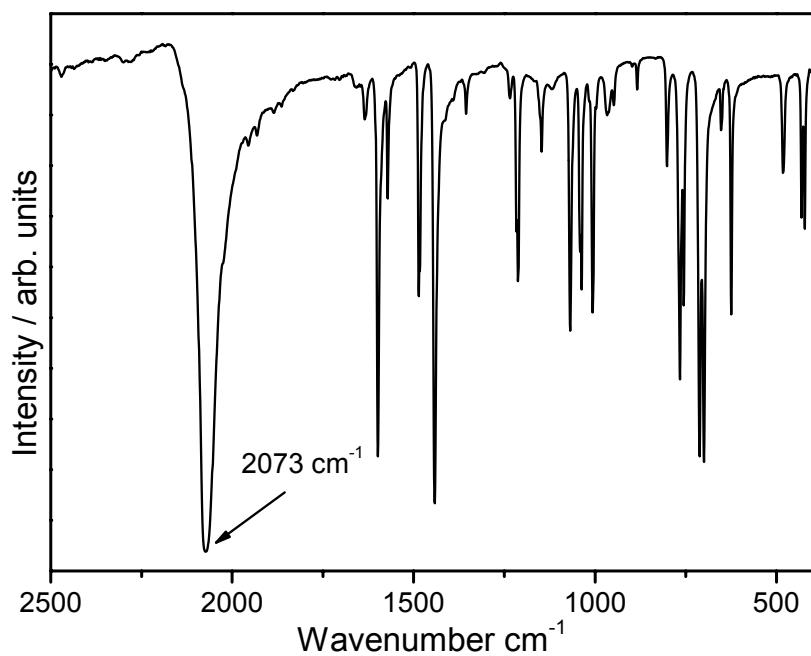


Fig. S1 IR spectra of compound $[\text{Co}(\text{SCN})_2(\text{pyridine})_4]$ (**1**).

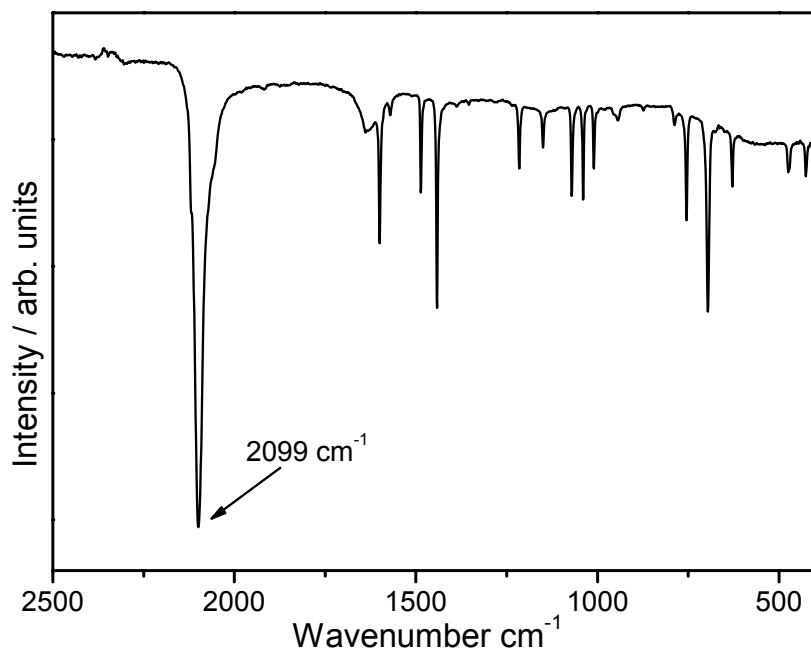


Fig. S2 IR spectra of compound $[\text{Co}(\text{SCN})_2(\text{pyridine})_2]_n$ (**2**).

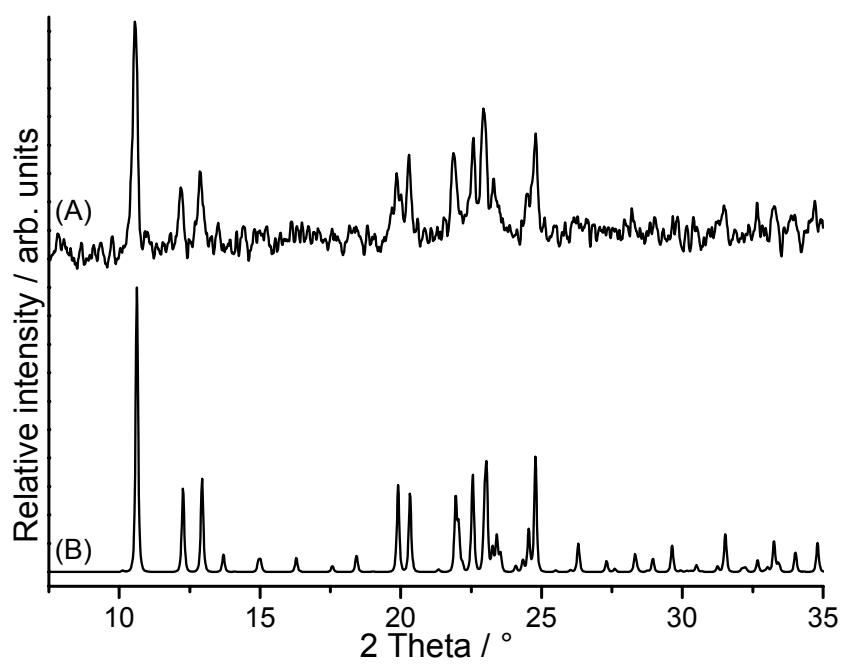


Fig. S3 Experimental XRPD pattern of compound $[\text{Co}(\text{SCN})_2(\text{pyridine})_4]$ (**1**) together with calculated XRPD pattern from single crystal data for compound **1**.

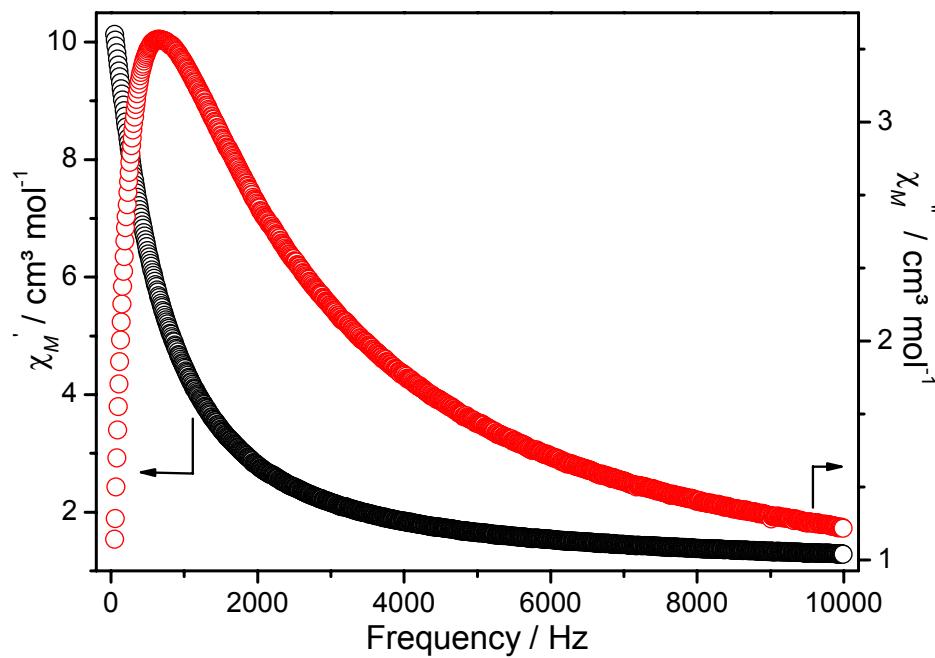


Fig. S4 Results of the magnetic measurements for compound $[\text{Co}(\text{NCS})_2(\text{pyridine})_2]_n$ (**2**) by plots of in-phase χ_M' (black) and out-of-phase χ_M'' (red) ac magnetic susceptibility as function of frequency.