Supplementary Information

$[Fe^{III}(dmphy)(CN)_4]^-$: a new building block for designing single-chain magnets

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Tabla S1. Selected ac magnetic data for **5**

T / K	$\chi_{T(A)}$ / $\chi_{T(B)}$ [a]	$\chi_{S(A)}$ / $\chi_{S(B)}$ [b]	$\alpha_{(A)}$ / $\alpha_{(B)}$ [c]	$-\log \tau_{(A)}$ / $-\log \tau_{(B)}$ [d]
2.0	2.08 / 11.82	0.08 / 0.90	0.445 / 0.641	2.26 / ----
2.3	2.88 / 13.50	0.05 / 1.20	0.423 / 0.621	2.98 / -----
2.5	3.72 / 12.00	0.01 / 2.00	0.422 / 0.530	3.34 / 0.23
2.7	6.00 / 17.10	0.00 / 3.00	0.420 / 0.490	3.70 / 0.88
3.0	5.50 / 13.70	0.00 / 3.60	0.325 / 0.440	----- / 1.80
3.3	----- / 19.00	----- / 6.50	------ / 0.386	----- / 2.80

(a) Isothermal and (b) adiabatic susceptibilities and (c) Cole-Cole parameter in the following equation:

$$\chi''(\chi) = \frac{\chi_T - \chi_S}{2\tan\left[(1+\alpha)\pi/2\right]} + \left\{ (\chi - \chi_S)(\chi_T - \chi) + \frac{(\chi_T - \chi_S)^2}{4\tan^2\left[(1+\alpha)\pi/2\right]} \right\}^{1/2}$$

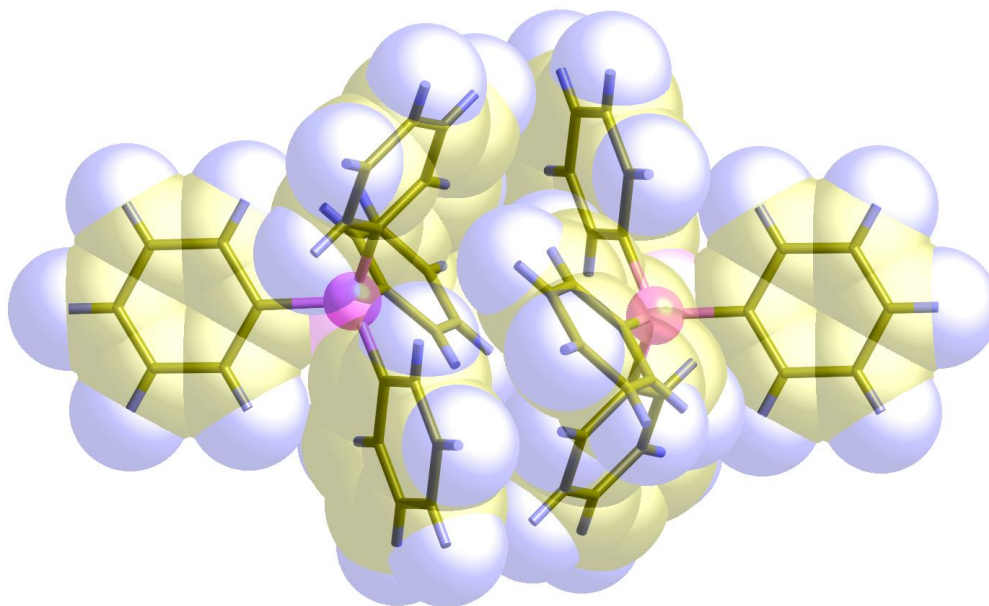(d) τ is the relaxation time at each temperature from this Table.

Figure S1. Sextuple phenyl embrace (SPE) interaction involving the PPh$_4^+$ cations in **1.**
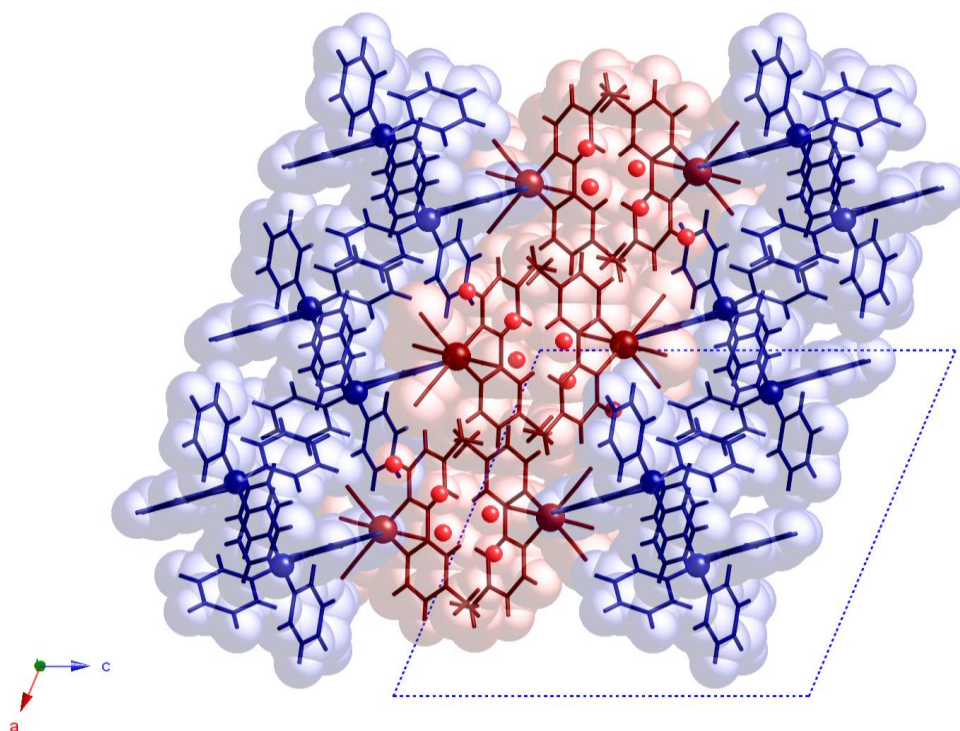
Figure S2. View in the *ac* plan of the double alternative packing of the PPh$_4^+$ cations and [Fe(dmbipy)(CN)$_4$]$^-$ anions in **1**.
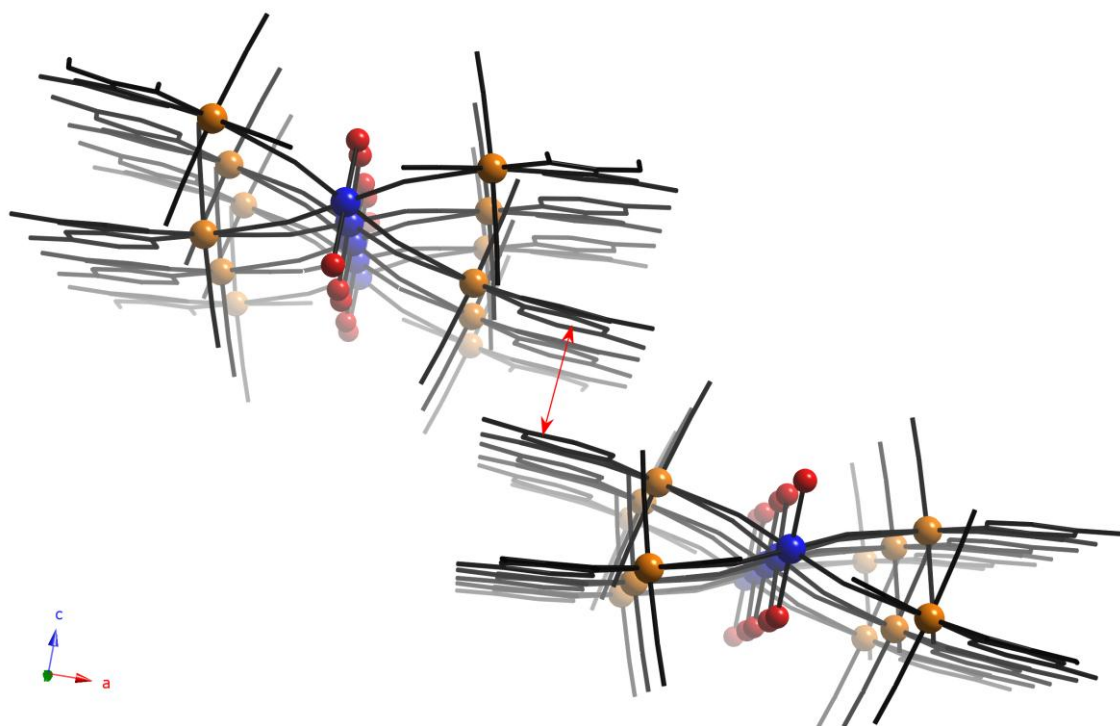
Figure S3. View of two neighbouring chains along the crystallographic *b* axis showing the shortest π-π interactions between the quasi-eclipsed dmbpy ligands.
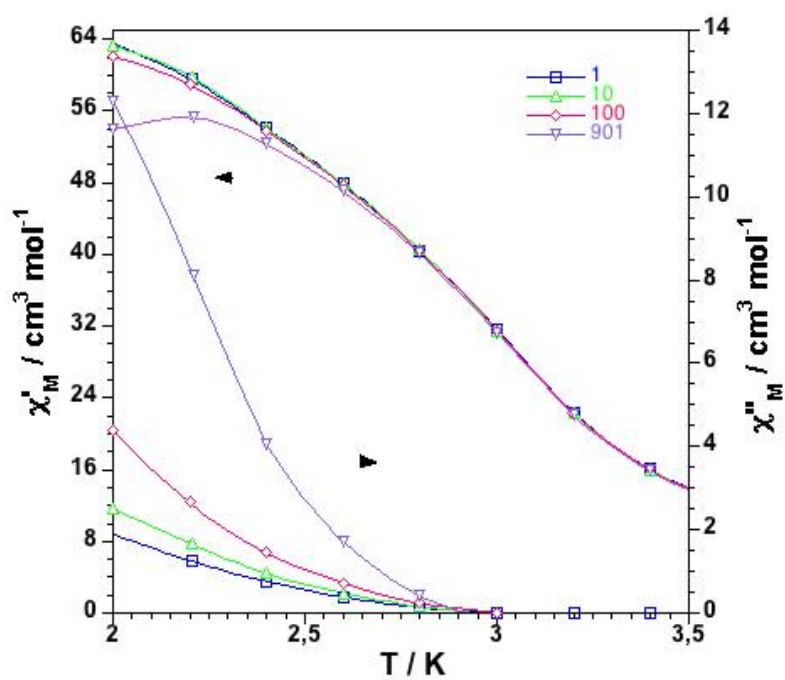
Figure S4. Temperature dependence of the *ac* susceptibility for **3** measured at several frequencies (1–901 Hz), where χ_M' and χ_M'' are in-phase and out-of-phase susceptibilities, respectively. The solid lines are eye-guides.
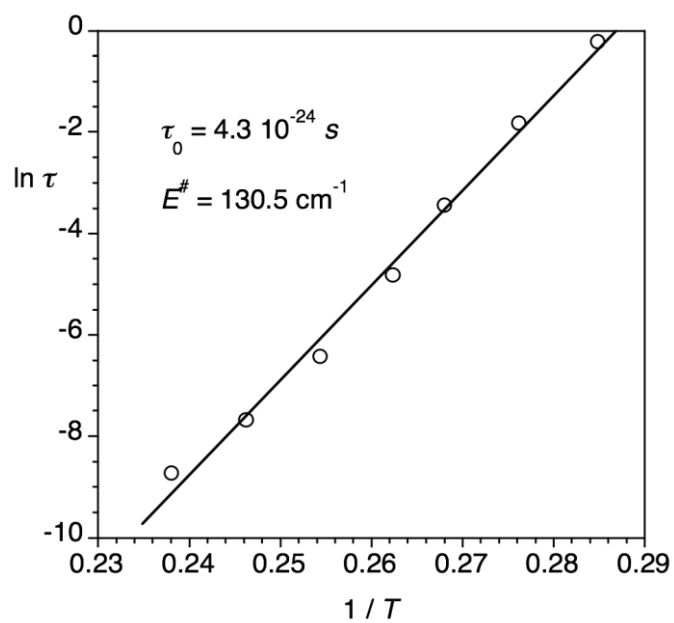
Figure S5. Arrhenius plot for **5**: (O) experimental data from de *ac* susceptibility (Figure 9); (——) least-squares fit to the Arrhenius law.
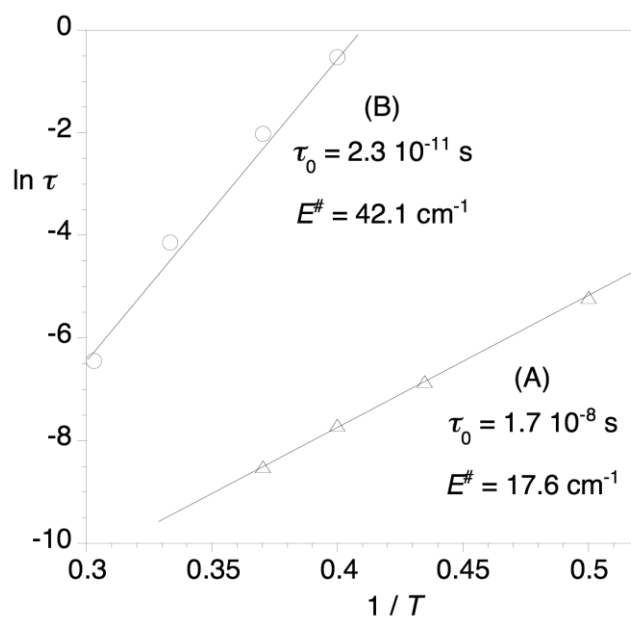
Figure S6. Arrhenius plots for **5**: (O) and (△): experimental data extracted from the Cole-Cole plots (Fig. 10) corresponding to the two different relaxation processes noted A and B (see text).