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A new approach to the synthesis of heteronuclear propeller-like SMMs

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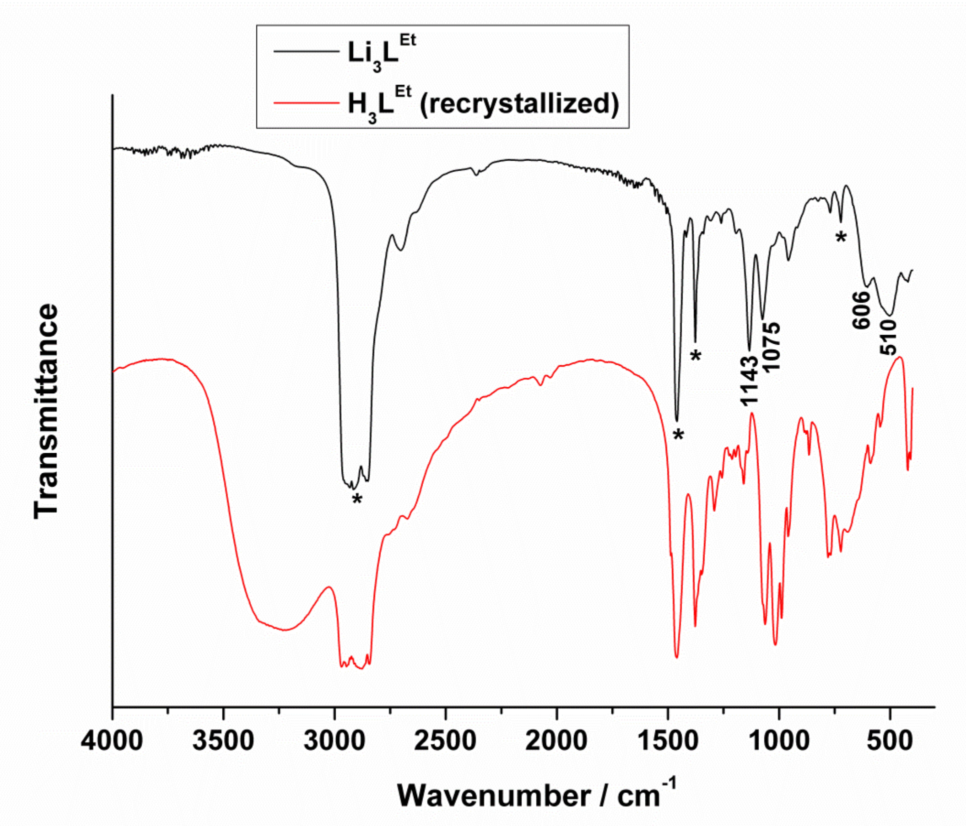

Fig. S1 Comparison of the FTIR spectra (Nujol mulls) of the tripodal alcohol (H_3L^{Et} = 2-hydroxymethyl-2-ethylpropane-1,3-diol, red line) and its corresponding lithiated alkoxide, Li_3L^{Et} (black line). Absorption bands for Nujol are marked with asterisks. In the spectrum of the alkoxide, the absence of the broad ν(O-H) band above 3000 cm^{-1} is indicative of full deprotonation of the corresponding alcohol.

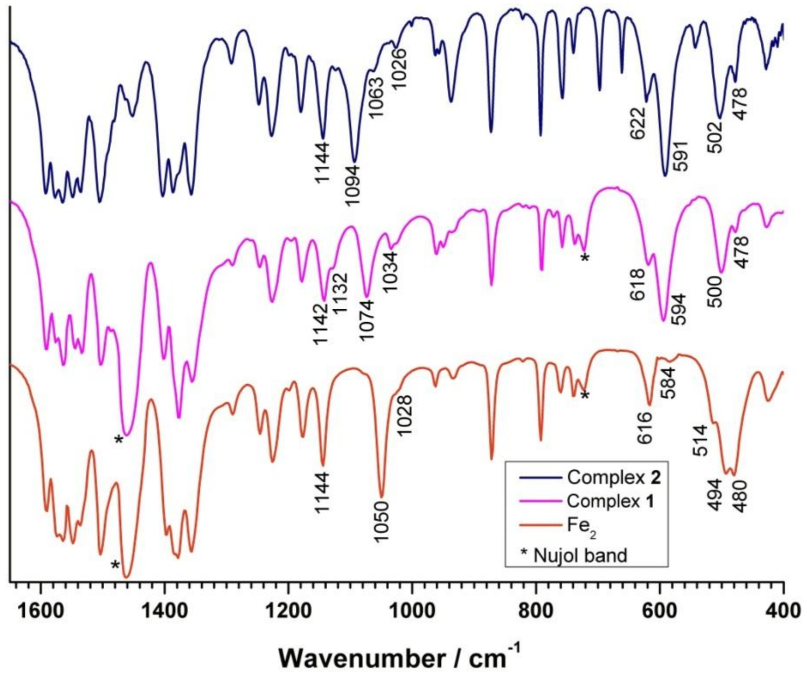

Figure S2 Comparison of the FTIR spectra of complexes **1** (pink line), **2** (blue line) and the dimeric iron(III) starting material, **Fe$_2$** (brown line), in the 400-1650 cm^{-1} region. The spectra of **Fe$_2$** and complex **1** were recorded in Nujol mulls, while the one for complex **2** was obtained from a KBr pellet. The most useful, diagnostic bands for the identification of the M$_3$M' topology are those assigned to ν(C-O$_{alkoxide}$) at 1074 (**1**) and 1094 cm^{-1} (**2**) and to ν(M-O$_{alkoxide}$) at 594 and 591 cm^{-1} for **1** and **2** respectively

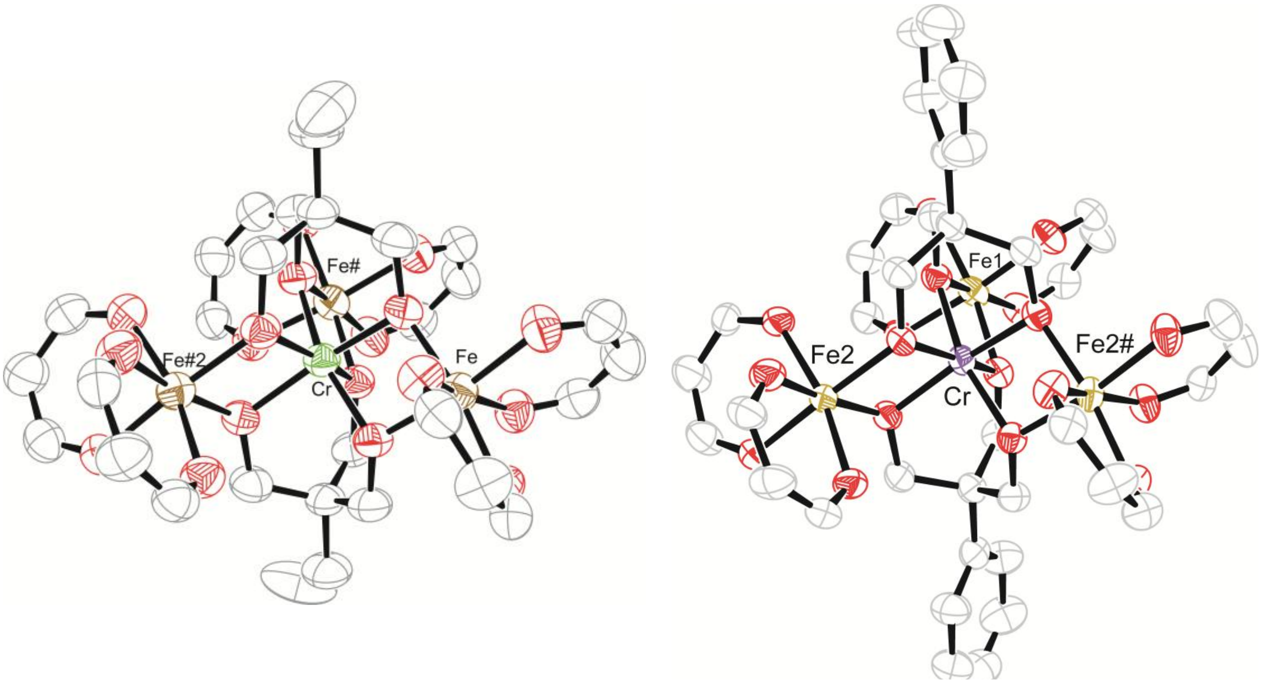

Fig. S3 ORTEP representations of the molecular structures of complex **1** (left) and **2** (right). Thermal ellipsoids are drawn at the 40% probability level. Bu^t groups of the dpm molecules are not shown for clarity.

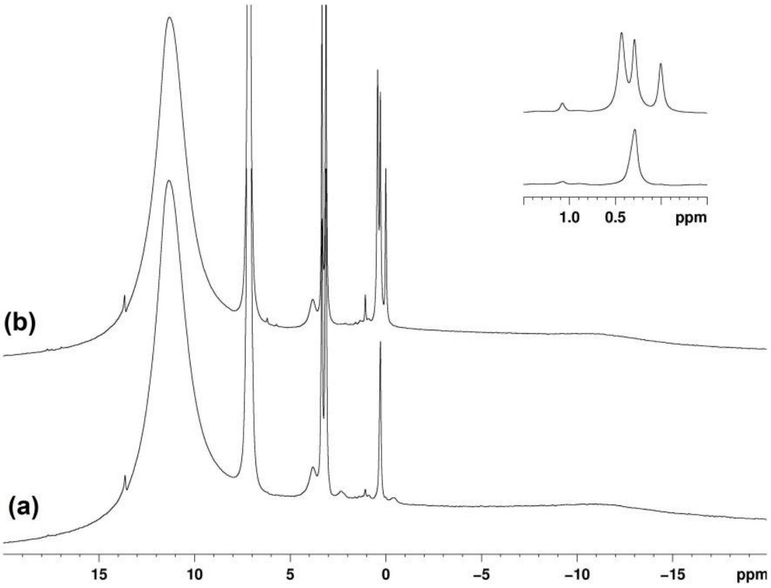

Fig. S4 ^{1}H-NMR spectra (400.13 MHz) recorded from a C_6D_6 solution of complex **2** at 303K. (a) Freshly prepared sample; (b) after 8 days at room temperature. The addition of tetramethylsilane (tms, $\delta = 0.0$ ppm) to the sample was carried out after recording spectrum (a). Inset: Low frequency region, evidencing the peaks at 1.07 (free Hdpm) and 0.44 ppm (possibly water). Note the very broad sinal at *ca.* -11.5 ppm, given by the methine resonance in Fe^{III}-bound dpm. The peak at *ca.* 13.7 ppm is a quadrature detection artifact at the center of the large spectral window employed in these experiments (+60 to –40 ppm).

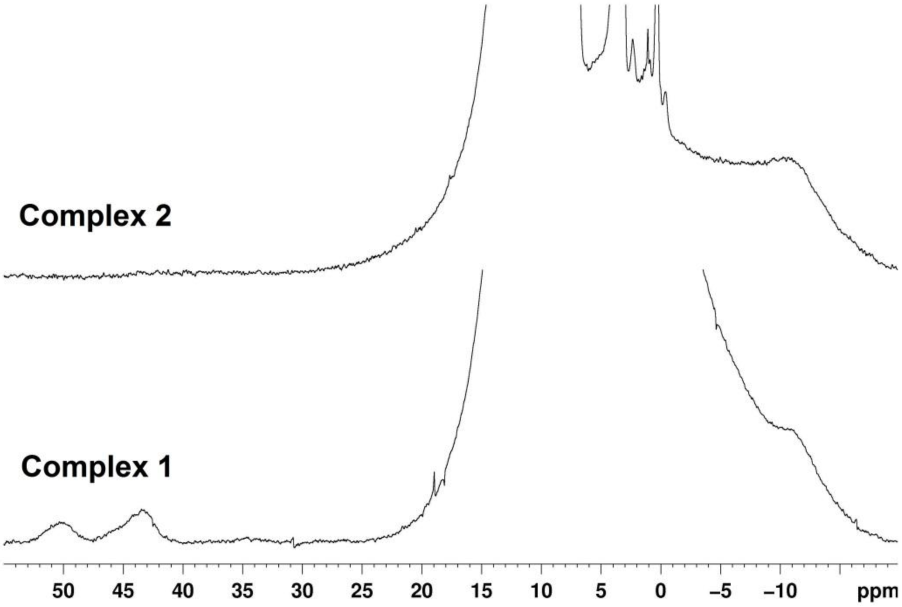

Fig. S5 Vertical expansion of the^{1}H-NMR spectra (400.13 MHz) recorded from C_6D_6 solutions of complexes **1** and **2** at 303K, emphasizing the low intensity, broad peaks tentatively assigned to the alkyl protons of the tripodal alkoxide in complex **1**, $[C\underline{\mathbf{H}}_3C\underline{\mathbf{H}}_2C(CH_2O)_3]^{3-}$ (43.2 and 50.3 ppm respectively), and to the methine hydrogen of dpm in both complexes [-11.0 (shoulder) and -11.5 ppm for **1** and **2** respectively].

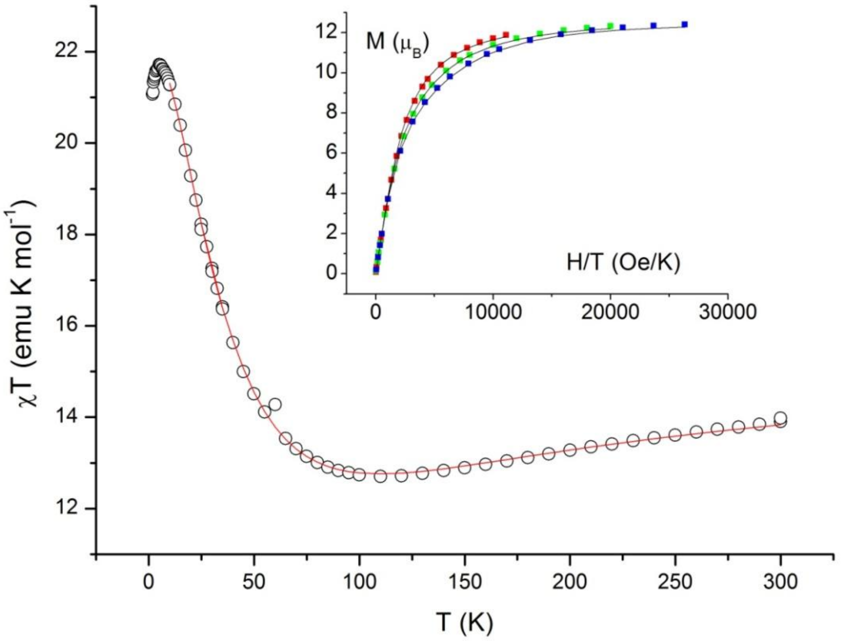

Fig. S6 Temperature dependence of the molar magnetic susceptibility multiplied by temperature of **2**. In the inset the field dependence of the magnetization at 1.9 (blue), 2.5 (green), and 4.5 K (red) versus the rescaled variable *H/T*. Solid lines represent the best fit values.

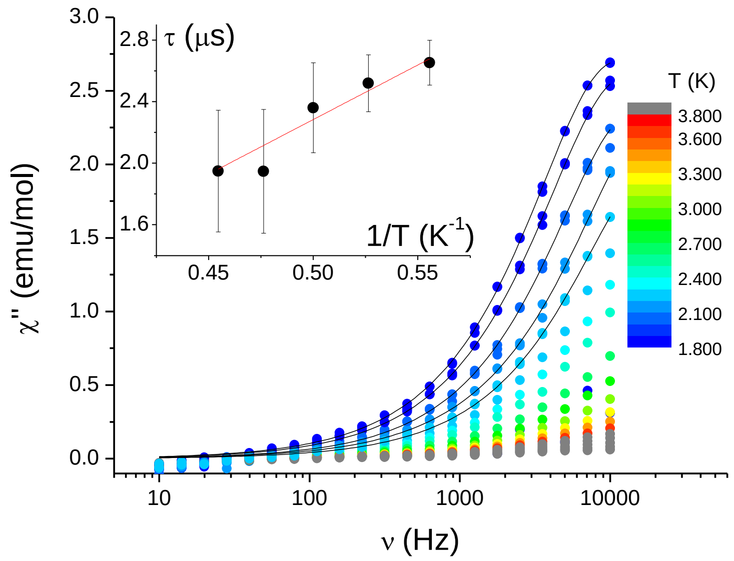

Fig. S7 Frequency dependence of the out-of-phase component of the molar ac susceptibility of **2** in H_{dc} = 1 kOe and several temperatures (see color legend). The black lines are the calculated values assuming one relaxing species. In the inset the temperature dependence of the relaxation time in a semi-logarithmic plot and in red the best fit curve obtained with the Arrhenius law.

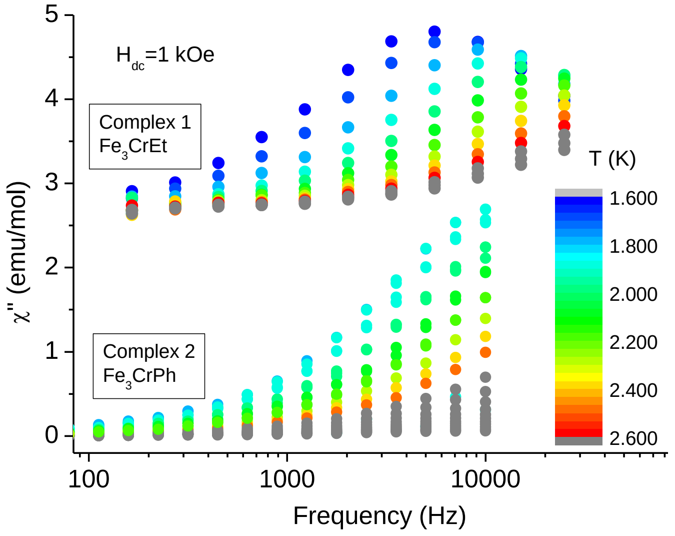

Fig. S8 Comparison of the out-of-phase component of the molar ac susceptibility for complexes **1** and **2** in H_{dc} = 1 kOe. The measurements are carried out with two different equipments. Data of 1 has been shifted vertically for clarity.

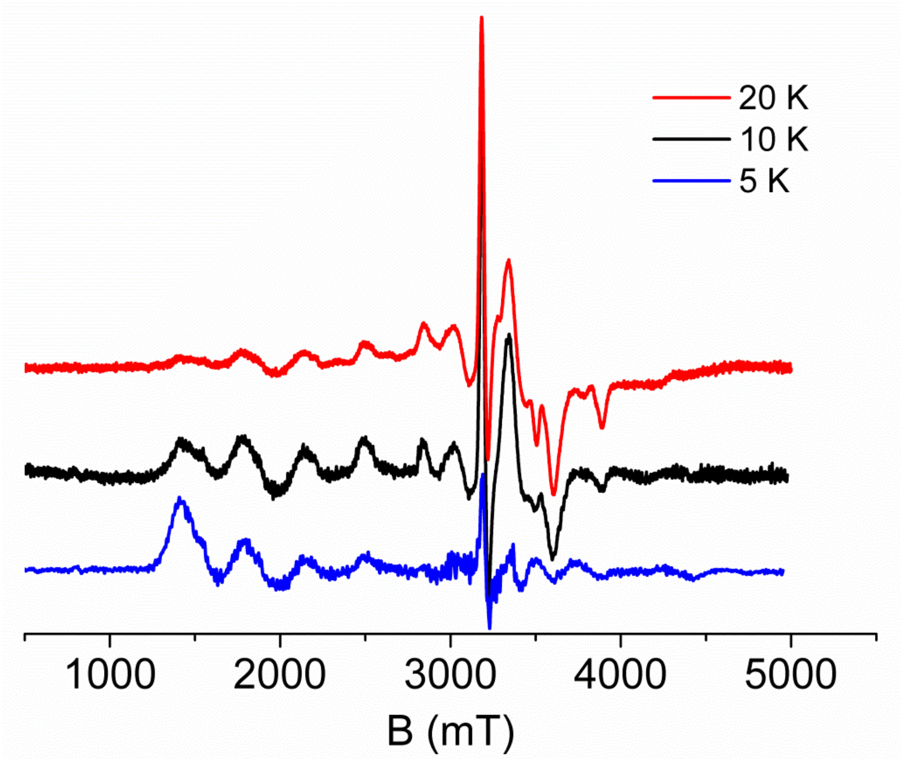

Fig. S9 Temperature dependence of the W-band (94.35 GHz) EPR spectrum of a microcrysllaine powder sample of **2**.