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Supplementary Information – New Journal of Chemistry 2014

Switching off the Single-Molecule Magnet properties of the [(Co^{II}(Me₆tren)(OH₂)]²⁺ module by complexation with *trans*-[Ru^{III}(salen)(CN)₂]⁻

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1. Electrochemical Experiments: Cyclic voltammetry measurements were performed using a CHI 760c potentiostat in a standard one-compartment cell under N_2 equipped with platinum wires for working/counter electrodes and silver wire (Ag/Ag^+) for the reference electrode. Solutions were prepared from 0.1 M tetraethylammonium tetrafluoroborate ([NEt₄⁺][BF₄⁻]) in acetonitrile. Ferrocene was added at the end of each experiment and potentials are referenced to the Cp₂Fe^{+/0} couple.



Figure S1. Cyclic voltammogram of complex 1, v = 100 mV/s.



Figure S2. Cyclic voltammogram of 2, v = 100 mV/s.



Figure S3. Cyclic voltammograms of *trans*-(PPh₄)[Ru^{III}(salen)(CN)₂], v = 100 mV/s; two different experiments: a) Anodic wave; b) Cathodic wave.

2. Optical Studies: Surface optical reflectivity measurements were performed on a home-built system at different temperatures ranging from 10 to 300 K. Heating and cooling rates were maintained at 4 K min⁻¹ during the measurements. The setup collects the light reflected by the sample (sum of direct and diffuse reflected light), which was analyzed by a high sensitivity Hamamatsu 10083CA spectrometer between 400 and 1000 nm. The spectra were compared to a white reference obtained with a NSIT traceable standard for reflectance (SphereOptics, ref SG3054). The background, acquired with the light source switched off, was subtracted from all measurements. The reflectivity can be plotted as a function of temperature, time or wavelength. In Figure S4, the thermal evolution of the absolute reflectivity was plotted at 480, 630 and 920 nm.



Figure S4. Absolute surface reflectivity of complex **2** at 480, 630 and 920 nm as a function of temperature with a temperature sweeping rate of 4 K/min under a spectroscopic white light ($P = 0.4 \text{ mW/cm}^2$)

3. *Magnetic Measurements:* The samples were checked by X-ray diffraction prior to any magnetic measurements. Magnetic susceptibility measurements were performed using Quantum Design MPMS-XL SQUID and PPMS-9 magnetometers. The measurements were carried out on freshly filtered polycrystalline samples introduced in a polyethylene bag ($3 \times 0.5 \times 0.02$ cm). dc measurements were conducted from 300 to 1.85 K and between ± 70 kOe applied dc fields. The thermal dependence of the magnetic susceptibility was measured at 1000 Oe. An *M* vs. *H* measurement was performed at 100 K to confirm the absence of ferromagnetic impurities. The *M* vs *H* data were also collected between 1.85 - 8 K while sweeping the magnetic field between 0 and 7 T at about 100 to 400 Oe/min. ac susceptibility experiments were realized at ac frequencies ranging from 10 to 10000 Hz with an ac field amplitude of 1 Oe (PPMS) and from 1 to 1500 Hz with an ac field amplitude of 3 Oe (MPMS). Experimental data were corrected for the sample holder and for the diamagnetic contribution of the samples.



Figure S5. M vs H plot of complex 1 (left) and 2 (right) in the temperature range from 1.85 to 8 K.



Figure S6. Frequency dependence of the in-phase (χ ', left) and out-of-phase (χ '', right) components of the ac susceptibility for **1** recorded at 1.9 K under different dc fields between 0 and 7000 Oe and with an 1–Oe ac field between 10 and 10000 Hz. Solid lines are guides for the eye.



Figure S7. Frequency dependence of the in–phase (χ ', left) and out–of–phase (χ '', right) components of the ac susceptibility for **2** recorded at 1.9 K under different dc fields between 0 and 10000 Oe and with an 1–Oe ac field between 10 and 10000 Hz. Solid lines are guides for the eye.