Supplementary Information



Figure S1. Temperature dependence of the magnetic moment normalized by weight for powders of compounds **1** and **2** at 1K/min heating/cooling rate. The different intensity of the HS paramagnetic signal is due to the higher Fe(II) density in **1** (molar mass = 722.5 g/mol) vs 2 (molar mass = 1671.5 g/mol).



Figure S2. AFM topography (left) and phase (right) images of the films of compound **1** cast from solutions at 100 °C (a) and at 20 °C (b) (see **Figure 2a**).



Figure S3. Diffuse reflection values at 400 nm as a function of film thickness from hot cast films of compound **1**. The red line is intended as a guide to the eye and emphasises the increasing degree of light scattering with increasing film thickness.



Figure S4. (a) Diffuse reflection values at 550 nm as a function of the thickness for films of compound **2** cast from fresh (at 100 °C and 20 °C) and gelled solutions. Scattering is found to be very low in films spin-coated from gels. (Lines are shown to guide the eye) (b) Diffuse reflection spectra recorded for a fused silica substrate (red line) and 780 nm thickness film of **2** (black line). (c) AFM topography (left) and phase (right) images of a 780 nm thickness film. Rms roughness is 15.8 nm.



Figure S5. Transmission UV-Vis spectra for a \sim 3 µm thickness dropcast film of **1** in the LS (blue, RT) and HS (red, > 90 °C) states, showing retention of the thermochromism associated with SCO.



Figure S6. (a) Transmission and reflection spectra in the UV-Vis range for a fused silica substrate (grey line) and 30 (black line), 65 (red line) and 90 (blue line) nm thickness films of compound **1** recorded at room temperature. Fabry-Perot interference fringes feature in both the T and R spectra. (b) Absorption from a 90 nm thickness film calculated as 1-R-T, showing the absorption centred at 535 nm characteristic of the LS state. The red line used as a baseline is obtained via non-linear fitting (Abs = $a + b/\lambda^2 + c/\lambda^4$). (c) Dependence of the absorbance at 535 nm on the film thickness and linear interpolation of the data (red line).

The absorbance (Abs) was calculated as

$$Abs = log[0][100/(R+T)]$$

where R and T are expressed as % values. Knowing the thickness, we can estimate the absorption coefficient (α) as directly proportional to the absorbance according to the Beer-Lambert expression; a linear fit of the absorbance at 535 as a function of thickness yields α .



Figure S7. ε' statistics from devices of different areas and film thickness for compound **1** (a) and **2** (b). Typical frequency response for ε' and ε'' for compound **1** (c) (device area: 0.02 cm², film thickness: 30 nm), compound **2** (d) (device area: 0.06 cm², film thickness: 395 nm) and comparison of ε' values for the two materials (e).



Figure S8. (a) Frequency dependence of $tan(\delta)$ at different temperatures on heating a 30 nm film of **1**. (b) Thermal hysteresis of $tan(\delta)$ at different frequency values for the same film. (c) Difference between the $tan(\delta)$ values measured on cooling and on heating, as a function of temperature at f = 100 Hz (red), f = 1 kHz (black) and f = 10 kHz (blue).



Figure S9. (a) Thermal hysteresis of $tan(\delta)$ at different frequency values for 395 nm thickness film of **2**. (b) Difference between the $tan(\delta)$ values measured on cooling and on heating at different frequency values.