

Supporting information

Spin crossover in Fe(triazole)-Pt nanoparticle self-assembly structured at the sub-5 nm scale

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Content: statement of contributions, supplementary experimental data.

Statement of contributions: All authors participated to the planning, discussed the results and contributed to the preparation of the manuscript. JSC and LS synthesized and characterized the ligand and the complex. ST, AG and MR worked on the Pt nanoparticle synthesis, self-assembly and characterization. ST, SU, AG, MM and BLK carried out XAS measurements. SU and JC carried out or analyzed magnetic and transport measurements.

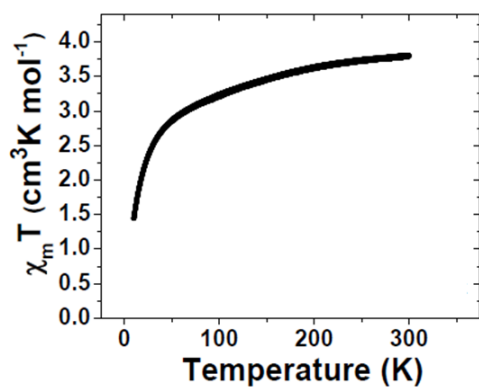


Fig. S1 Loss of spin crossover in the ‘powder state’. Magnetic measurement on the FeL_3 coordination polymer in the dry ‘powder state’. The curve is normalized per mole of Fe. The decrease of $\chi_m T$ at low temperature is attributed to zero-field splitting.

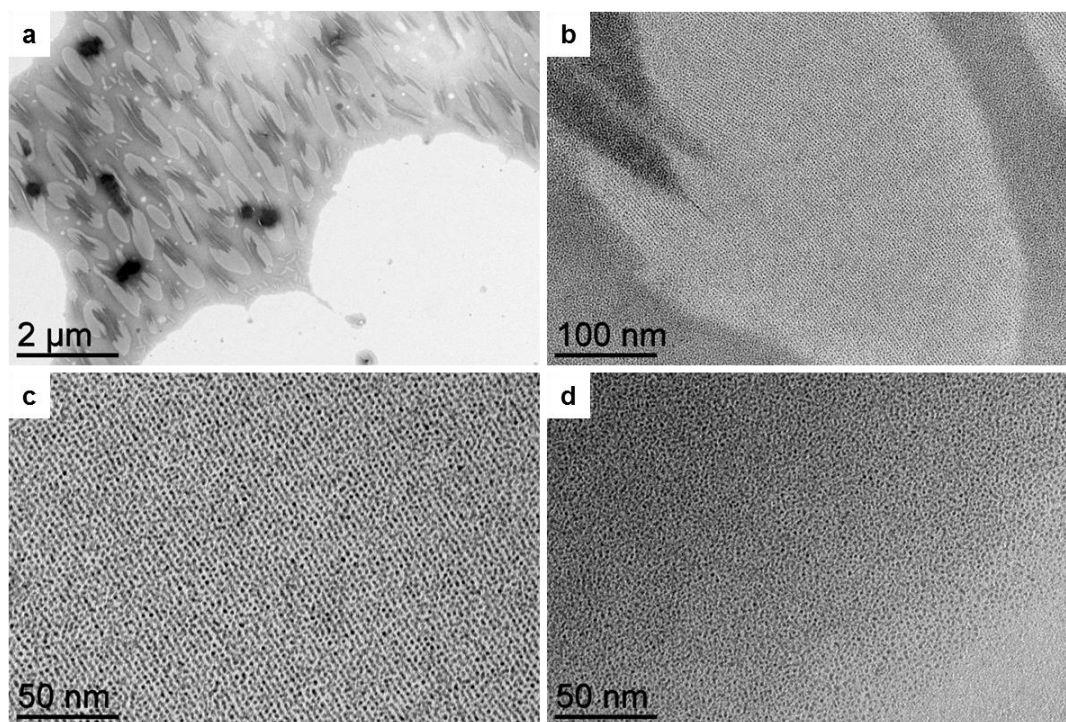


Fig. S2 Hybrid self-assembly made of Pt nanoparticles (NPs) and FeL_3 coordination polymers. (a, b, c) TEM images of the self-assembly at different magnifications (Fig. S2c is identical to Fig. 2a). The hybrid system is constituted by zones of NP lines distributed in larger areas where the PtNPs are disorganized. (d) TEM image after mixing of PtNPs with **L** only. No NP line is observed in absence of Fe(oTs)_2 .

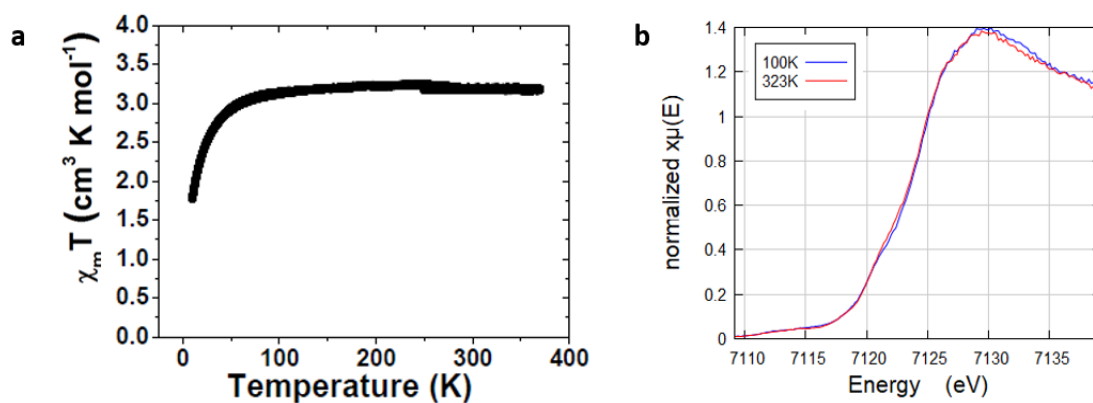


Fig. S3 Loss of spin crossover of the PtNPs-FeL₃ self-assembly in the ‘powder state’: (a) Magnetic measurement. The curve is normalized per mole of Fe. The sample stays high spin at all temperatures; the decrease of $\chi_m T$ at low temperature is attributed to zero-field splitting. (b) XAS measurement. The curves are identical at low and high temperature.

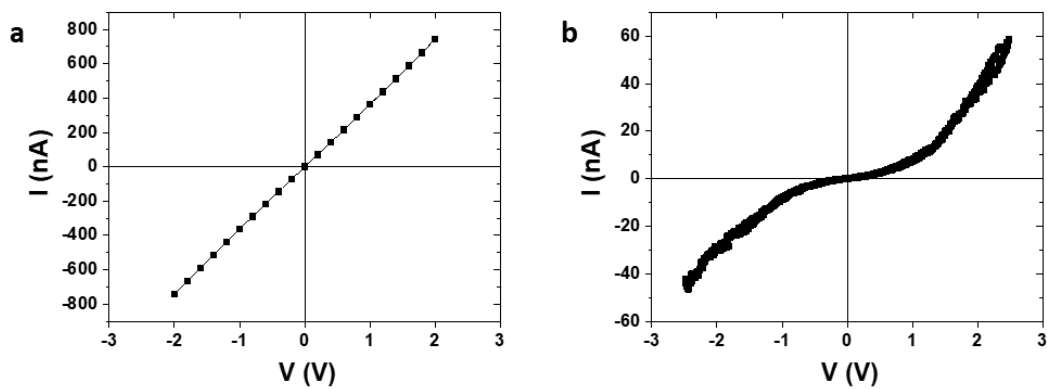


Fig. S4 I-V characteristics at 300K: (a) on a deposit of the pristine PtNPs; (b) on the PtNPs-FeL₃ self-assembly. The non-linearity of the curve for the self-assembly is the signature of the presence of Coulomb blockade (see *e.g.* S. Tricard, O. Said-Aizpuru, D. Bouzouita, S. Usmani, A. Gillet, M. Tassé, R. Poteau, G. Viau, P. Demont, J. Carrey and B. Chaudret, *Mater Horiz*, 2017, **4**, 487–492).

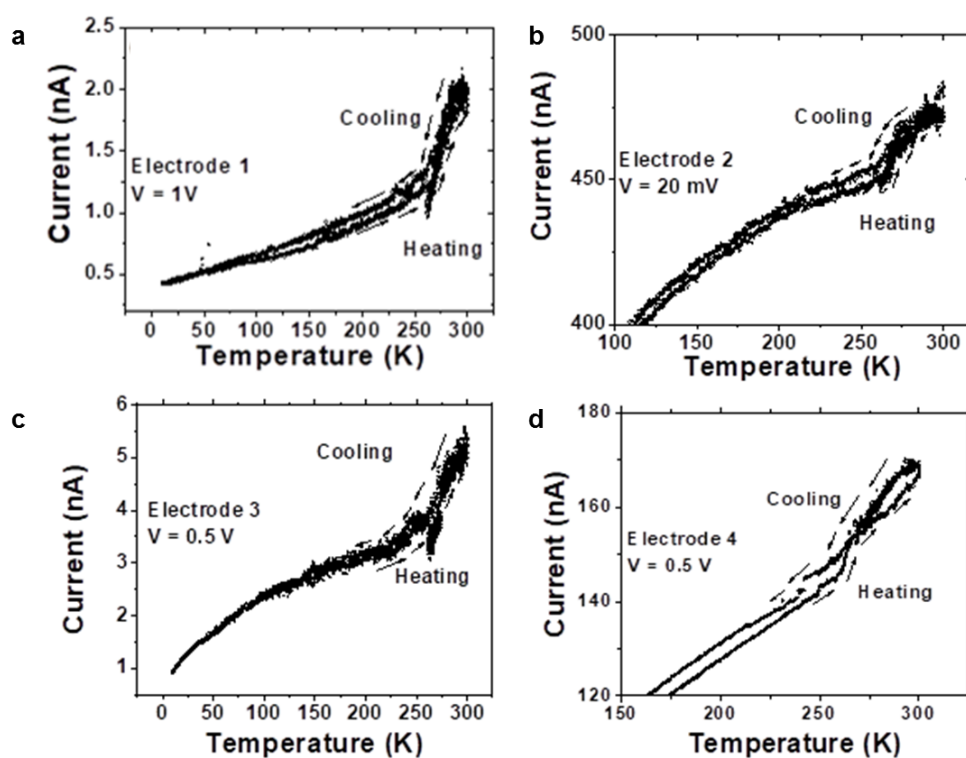


Fig. S5 Electrical measurements on the PtNPs-FeL₃ self-assembly on independent electrodes, biased at different voltages: (a) 1V; (b) 20 mV; (c) 0.5V; (d) 0.5 V, with sweeping rate of 1K.min⁻¹. All the curves slope breaks around 270 K can be attributed to the spin crossover. The significant difference in resistance between the electrodes is attributed to non-homogeneous filling of the interdigitated electrodes by the materials.

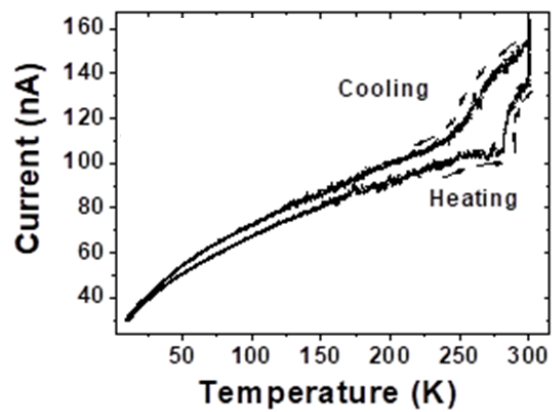


Fig. S6 Hysteresis opening in electrical measurements on the PtNPs-FeL₃ self-assembly, on electrodes biased at voltage of 0.1V, with a sweeping rate of 3 K.min⁻¹.