Supplementary information

for

Synthesis, characterisation and magnetic properties of octahedral chromium(III) compounds with six C-donor ligands


- **Figures S1–S3:** Simulation of EPR spectra registered for [NBu₄][trans-Cr(C₆F₅)₄(CN'Bu)₂] (2) at representative stages of the observed evolution Pages S1–S3

- **Figure S4:** EPR spectra of [NBu₄][trans-Cr(C₆F₅)₄(CN-Xy)₂] (3) compared with typical spectra of 2 Page S4

- **Figure S5:** Bulk magnetic properties of 3 Page S5
**Figure S1.** Comparison between experimental (red lines) and simulated (blue lines) EPR spectra of a polycrystalline sample of 2 measured at X-band (upper part) and at Q-band (lower part). Experimental spectra correspond to those given in Figures 5c (X-band) and 6c (Q-band). Simulated spectra correspond to type C contribution (Table 3).
Figure S2. Comparison between experimental (red lines) and simulated (blue lines) EPR spectra of a polycrystalline sample of 2 measured at X-band (upper part) and at Q-band (lower part). Experimental spectra correspond to those given in Figures 5d (X-band) and 6d (Q-band). Simulated spectra have been calculated considering coexistence of type C (80%) and type D (20%) contributions (Table 3).
Figure S3. Comparison between experimental (red lines) and simulated (blue lines) EPR spectra of a polycrystalline sample of 2 measured at X-band (upper part) and at Q-band (lower part). Experimental spectra correspond to those given in Figures 5e (X-band) and 6e (Q-band). Simulated spectra correspond to type D contribution (Table 3).
**Figure S4.** EPR spectra of 3 (red solid line) compared with typical spectra of 2 (black broken line): a) X-band; b) Q-band.
Figure S5. a) Magnetization vs. the reduced applied magnetic field, $M(\mu_0H/k_B T)$, of a powder sample of 3 at 1.8 K (dotted line corresponds to the evolution calculated with a Brillouin function with $S = 3/2$ and $g = 1.92$); b) Thermal dependence of the magnetic susceptibility, $\chi$ (empty circles, ordinate scale on the left) and its inverse, $\chi^{-1}$ (full circles, ordinate scale on the right) of the same sample (dotted lines correspond to the predicted evolution following the Curie law).