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Structural and magnetic studies of mononuclear lanthanide complexes derived from N-rich chiral Schiff bases.

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Fig. S1. Powder X-Ray spectra for the diluted complexes **2d** and **3d**. In black is represented the simulated powder spectra from the single crystal structure of the complex **2**.



Fig. S2. IR spectra for complexes [Ce(L)(NO₃)₃(MeOH)] (**1**), [Gd(L)(NO₃)₃] (**2**) and [Dy(L)(NO₃)₃] (**3**) the diluted **2d** and **3d**.

Table S1. CShM values for the undecacoordinated environment of complex **1** and the decacoordinated complex **2**. The relatively high distortion with the ideal polyhedra are due to the low bite of the nitrato and *cis* N-donors of the Schiff base.

11-vertex Polyhedra	Ce ^{III} (1)	10-vertex Polyhedra	Gd ^{III} (2)
Capped pentagonal	5.64	Bicapped cube (D4h)	9.77
prism (C5v)			
Capped pentagonal	5.99	Bicapped square	4.67
antiprism (C5v)		antiprism (D4d)	
Augmented	7.73	Metabidiminished	7.08
sphenocorona (cs)		icosahedron (C2v)	
		Sphenocorona (C2v)	3.05
		Staggered	4.66
		Dodecahedron (D2)	



Fig. S3. $\chi_M T$ (left) and magnetization (right) plots for referred complexes. The $\chi_M T$ values at room temperature are very close to the expected values for a lonely isolated lanthanide ions (**expected**/experimental cm³·mol⁻¹·K): Ce^{III}, (²F_{5/2}, g = 6/7, **0.80**/0.74 cm³·mol⁻¹·K), Dy^{III} (⁶H_{15/2}, g = 4/3, **14.2**/13.8 cm³·mol⁻¹·K).



Fig. S4. *X*-band EPR spectra for the Gd^{III} compounds **2** (red) and **2d** (black). Reduction of dipolar interactions allows shaper lines for **2d**. The identical spectra confirm the same environment after dilution.



Fig. S5. χ_M " dependence of the transverse field for complex **1** (left) and **3** (right). From this initial dependence the fields to perform the *ac* measurements were 2000 G for **1** and 1000 G for **3**.



Fig. S6. χ'_{M} vs. *T* (left) and χ''_{M} vs. frequency (right) for complex **1**.



Fig. S7. Plot of $\ln(1/2\pi\nu)$ vs. T^{-1} from the $\chi''_{M}(T)$ data (red) and $\ln(\tau)$ vs. T^{-1} from the fit of the Argand plot (blue) for complex **1**.