Supporting Information

Slow relaxation of magnetization in lanthanide-biradical complexes based on a functionalized nitronyl nitroxide biradical

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Fig. S8 $M$ versus $H$ plot at 2 K for complex 3-Dy.

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Fig. S12 The $\tau$ versus $H$ plot for complex 3-Dy at 2.0 K under the applied dc field.
Table S1. Selected bond lengths [Å] and angles [°] for 1-Gd.

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Table S3. Selected bond lengths [Å] and angles [°] for 3-Dy.

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**Table S4.** SHAPE analysis for the Ln coordination spheres.

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SAPR-8: Square antiprism; TDD-8: Triangular dodecahedron; JBTPR-8: Biaugmented trigonal prism J50; BTPR-8: Biaugmented trigonal prism; JSD-8: Snub diphenoïd J84.

**Table S5.** Distances (Å) for the hydrogen bonds in all compounds.

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**Fig. S1** Molecular structure of 1-Gd and coordination polyhedrons of the Gd(III) ions (H and F atoms are omitted for clarity).

**Fig. S2** Molecular structure of 2-Tb and coordination polyhedrons of the Tb(III) ions (H and F atoms are omitted for clarity).
Fig. S3 Hydrogen bond-linked motifs in 1-Gd (partial H and all F atoms are omitted for clarity).

Fig. S4 Hydrogen bond-linked motifs in 2-Tb (partial H and all F atoms are omitted for clarity).

Fig. S5 Powder X-ray diffraction patterns of all complexes.
**Fig. S6** Field-dependent magnetization at 2.0 K for 1-Gd. The red line was simulated by the PHI program using the obtained magnetic parameters by the modeling of susceptibility data.

**Fig. S7** $M$ versus $H$ plot at 2 K for complex 2-Tb.

**Fig. S8** $M$ versus $H$ plot at 2 K for complex 3-Dy.
Fig. S9 Temperature dependence of the in-phase and out-of-phase components of the ac magnetic susceptibility in zero field with an oscillation 3 Oe for complex 2-Tb.

Fig. S10 Temperature-dependent ac signals under a zero dc field for 3-Dy.
Fig. S11 Frequency dependence of the $\chi'$ (top) and $\chi''$ (bottom) components of the ac susceptibility, between 0 and 5000 Oe and between 10 and 10000 Hz, for 3-Dy at 2 K.

Fig. S12 The $\tau$ versus $H$ plot for complex 3-Dy at 2.0 K under the applied dc field.