







$^{164}\text{Dy}_{0.07}\text{Y}_{0.93}(\text{m})$  the hysteresis loops open also in field below 3.5 K (Figs. 3b and S26) at slightly higher temperature than  $\text{Dy}(\text{t})$  and  $\text{Dy}(\text{m})$ . However, the minimization of the internal field also opens the loops at zero field.

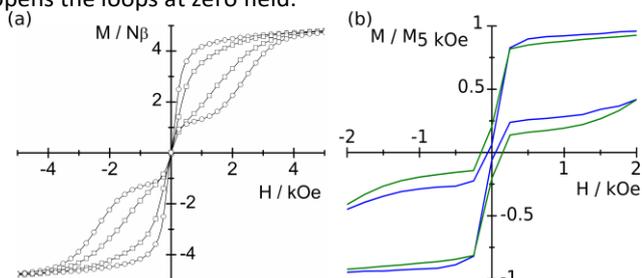


Fig. 3 (a) Hysteresis loops for (a)  $\text{Dy}(\text{t})$  (circles) and  $\text{Dy}(\text{m})$  (squares) for (b)  $^{164}\text{Dy}_{0.07}\text{Y}_{0.93}(\text{m})$  (olive) and  $\text{Dy}_{0.02}\text{Y}_{0.98}(\text{m})$  (blue) recorded at 500 mK at  $16 \text{ Oe s}^{-1}$ .

At  $^3\text{He}$  temperature (500 mK) the isotopically enriched material  $^{164}\text{Dy}_{0.07}\text{Y}_{0.93}(\text{m})$  is the best magnet (Fig. 3b), as expected. The remnant magnetization is multiplied by a factor of three with the isotopic enrichment. Finally,  $^{164}\text{Dy}_{0.05}\text{Eu}_{0.95}(\text{t})$  displays similar hysteresis (Fig. S27) than  $^{164}\text{Dy}_{0.07}\text{Y}_{0.93}(\text{m})$  which opens below 3.5 K. There is no striking difference between the hysteresis loops of these two compounds whatever the temperature (Fig. S28). The differences observed in condensed pure crystalline phase are not valid anymore once the molecule is dispersed in a crystalline diamagnetic medium. This confirms that a large part of the relaxation of the molecular magnetic moment at extremely low temperature is governed by the coupling with the surrounding moments.

In conclusion, a series of mononuclear coordination complexes of formula  $[\text{Ln}(\text{tta})_3(\text{L})] \cdot x\text{CH}_2\text{Cl}_2$  ( $x=0$  or  $1$ ) highlights two polymorphs depending of the lanthanide radii. The two  $\text{Dy}^{\text{III}}$  polymorphs behave as SMMs with an opened hysteresis loop at low temperature. Such magnetic bistability can be generally attributed to the combination of the  $\text{N}_2\text{O}_6$  environment with  $\text{Dy}(\text{tta})_3$  unit. The triclinic form presents a slower magnetic relaxation than its monoclinic analogue. Unambiguously, the symmetry of the coordination polyhedron plays a major role to determine the energy splitting diagram of the ground state multiplet. However, the electronic distribution at the surface of the coordination sphere of the lanthanide is at least as important as symmetry considerations. Finally the magnetic properties have been enhanced using magnetic dilution and isotopic enrichment in nuclear spin free Dysprosium which support the importance of isotopes for the control of quantum magnets.

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## Notes and references

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