## **Electronic supplementary information**

## Heteronuclear Ni(II)-Ln(III) (Ln = La, Pr, Tb, Dy) complexes: Synthesis and Single-Molecule Magnet Behaviour

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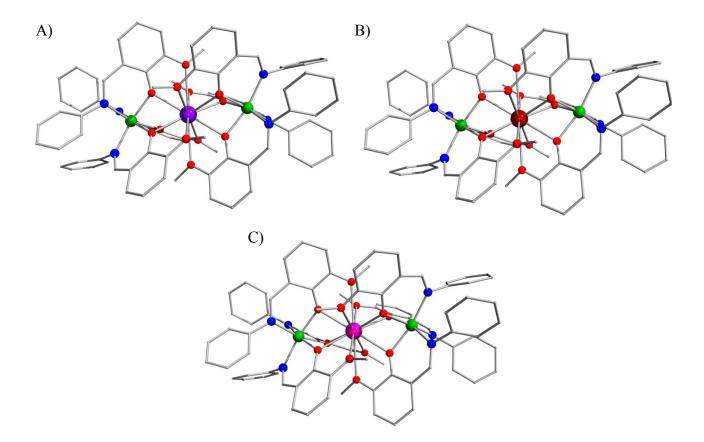


Figure S1. Crystal structures of cationic complexes of 1 (panel A), 2 (panel B) and 3 (panel C).

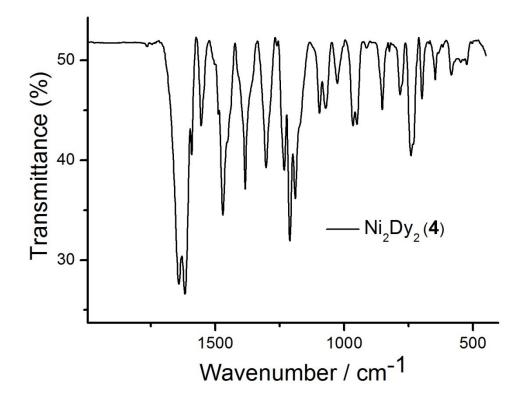
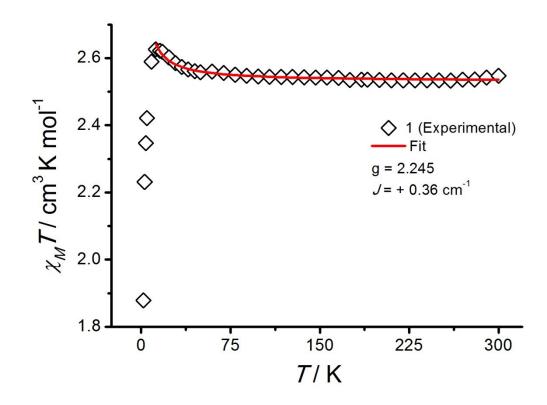
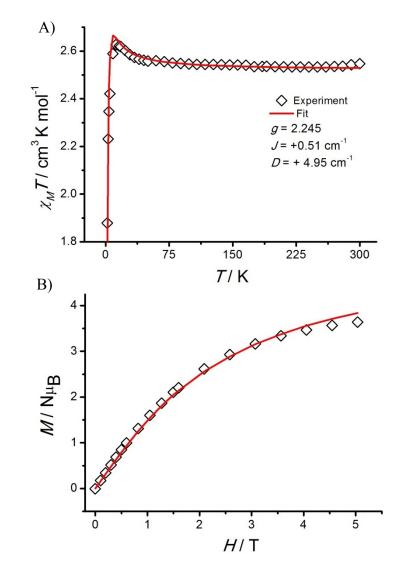


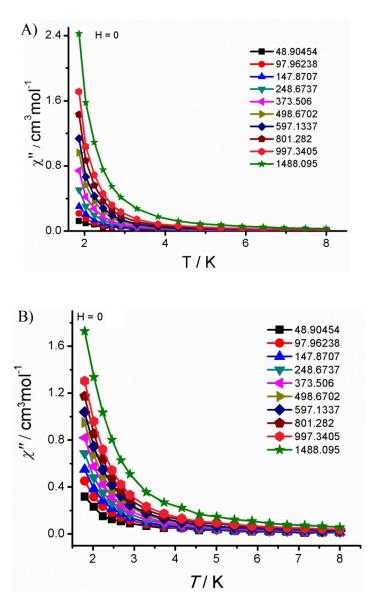
Figure S2. Infrared spectrum (KBr pellet) of complex 4.



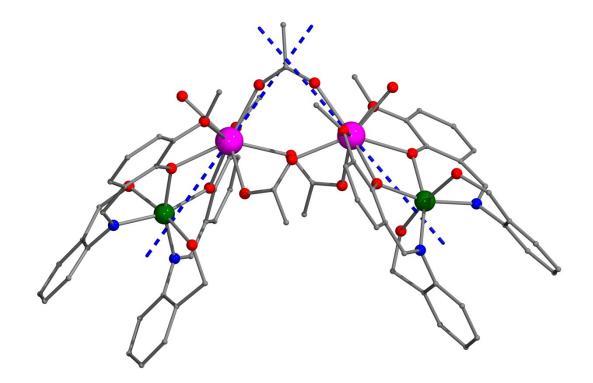
**Figure S3**. Temperature dependent magnetic susceptibility data ( $\chi_M T(T)$ ) of complex **1** measured in the presence of 0.1 Tesla. The red solid indicates the best fit obtained for the parameters (*g* and *J* without magnetic anisotropy) described in the plot.



**Figure S4**. A) Temperature dependent magnetic susceptibility data ( $\chi_M T(T)$ ) of complex **1** measured in the presence of 0.1 Tesla. B) Field dependent magnetization measurement of complex **1** measured at 2.0 K. The red solid indicates the best fit (simultaneous fit of  $\chi_M T(T)$  and M(H) data) obtained for the parameters (*g* and *J* with magnetic anisotropy) described in the plot.



**Figure S5**. Frequency dependent out-of-phase susceptibility signals for complexes **3** (panel A) and **4** (panel B) measured at the indicated frequency in the absence of external magnetic field.



**Figure S6**. Crystal structure of cationic complex of  $\{Ni_2Dy_2\}^{3+}$  is shown which is reported by us elsewhere. The dotted blue line represents the  $g_z$  orientation of the two Dy(III) ions derived from the electrostatic model (refer to main manuscript for details).