Electronic Supplementary Information

A rational approach to the modulation of the dynamics of the magnetization in a Dy - Nitronyl Nitroxide Radical complex

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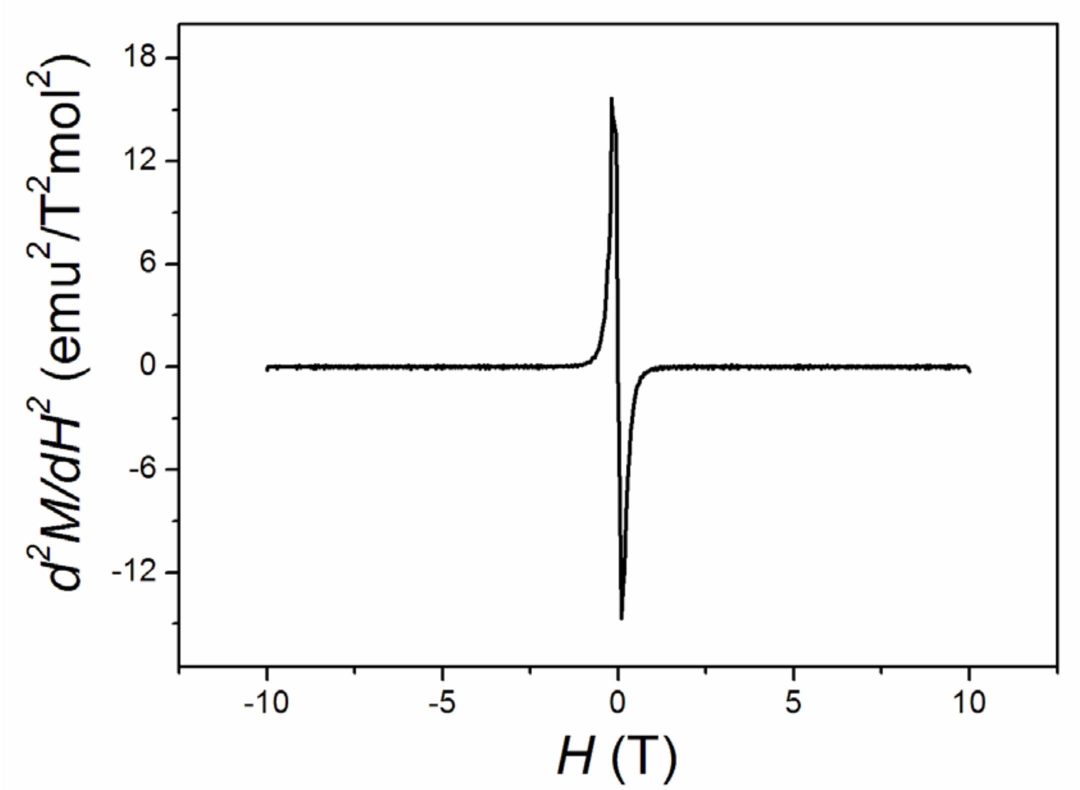

Fig. **S1**: Second derivative of the magnetization *vs* field curve, measured from 120 to -120 kOe at 1.5 K on a pellet sample.

Distribution function for the $[DyNITpPy]_2$ species, calculated with the Hamiltonian described in the text:

$$Z = 4\cosh(\beta G\mu_B H^z) + 4\cosh(\beta G\mu_B H^z) + 4\cosh(\beta\frac{(J_1+J_2)}{2}) + 2e^{\beta\frac{(J_1+J_2)}{2}}\cosh[(\beta(G+g)\mu_B H^z)]$$
$$+2e^{-\beta\frac{(J_1+J_2)}{2}}\cosh[(\beta(G-g)\mu_B H^z)]$$

The symbols bear the meanings pointed out in the main paper and $\beta = (k_B T)^{-1}$. With this function an evaluation of the $M(H,T)$ function has been achieved, based on the statistical relation $M = k_B T\frac{\delta\ln(Z)}{\delta H} = k_B T\frac{1}{Z}\frac{\delta Z}{\delta H}$:

$$M = k_B T\frac{1}{Z}\left\{2e^{\frac{\beta(J_1+J_2)}{2}}\beta(G+g)\mu_B\sinh\left[(\beta(G+g)\mu_B H^z)\right] + 2e^{-\frac{\beta(J_1+J_2)}{2}}\beta(G-g)\mu_B\sinh\left[(\beta(G-g)\mu_B H^z)\right] + \right.$$
$$\left. +4\beta g\mu_B\sinh(\beta g\mu_B H^z) + 4\beta G\mu_B\sinh(\beta G\mu_B H^z)\right\}$$

Hence the temperature dependence of $\chi_{static}(= M/H)$ followed; for a reference see Kahn, O., *Molecular Magnetism*, VCH, 1993.

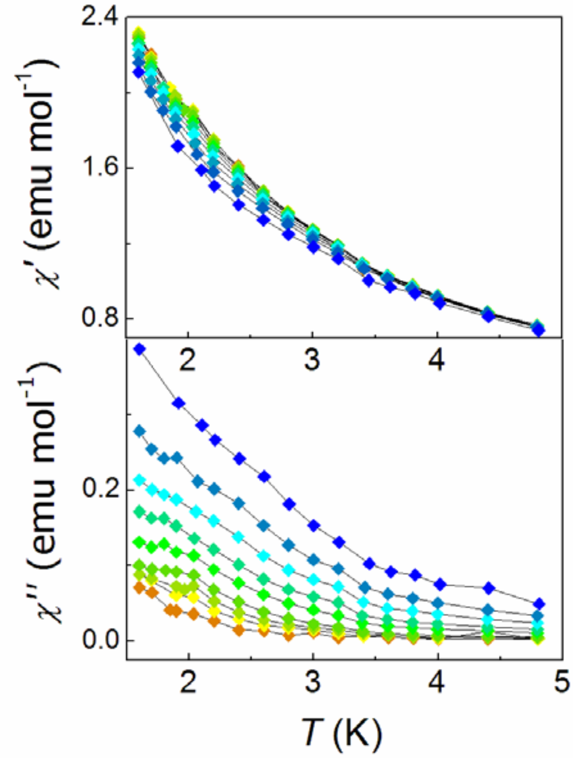

Fig. **S2**: χ' (top) and χ'' (bottom) *vs* T diagrams for a polycrystalline sample of the doped $[Dy\text{-}YNITpPy]_2$ species, measured in zero external field from 1.6 to 4.8 K using ten logarithmically spaced frequencies, ranging from 100 Hz to 25 kHz.

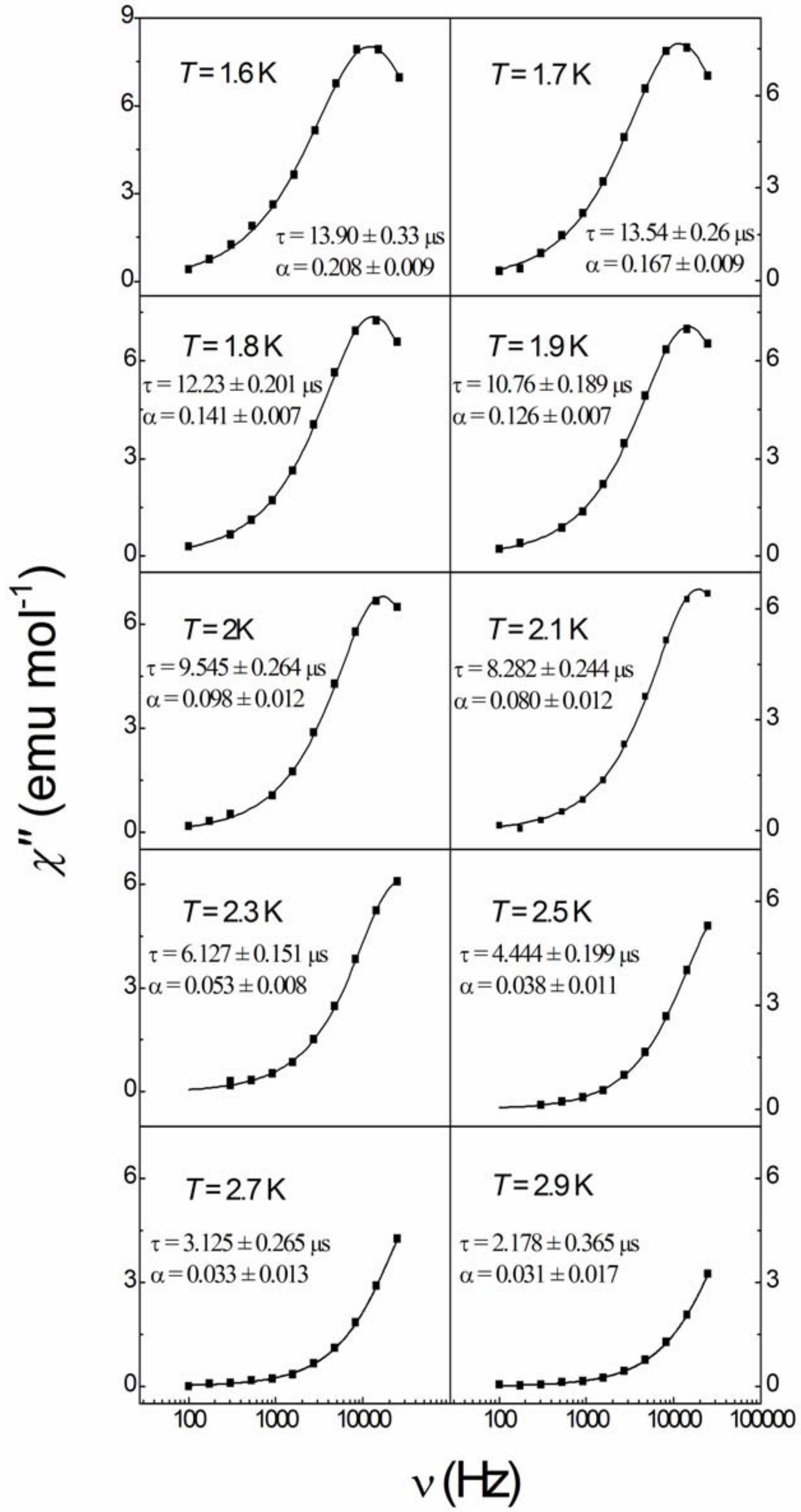

Fig. **S3**: χ'' *vs* $\log\nu$ diagrams extracted from *ac* susceptibility measurement of $[DyNITpPy]_2$, taken in zero external magnetic field for ten different temperatures ranging from 1.6 to 2.9 K. Solid lines: fits obtained with the following expression:[16]

$$\chi''(2\pi\nu) = \left(\chi_T - \chi_S\right)\frac{(2\pi\nu\tau)^{1-\alpha}\cos(\pi\alpha/2)}{1+2(2\pi\nu\tau)^{1-\alpha}\sin(\pi\alpha/2)+(2\pi\nu\tau)^{2-2\alpha}}$$

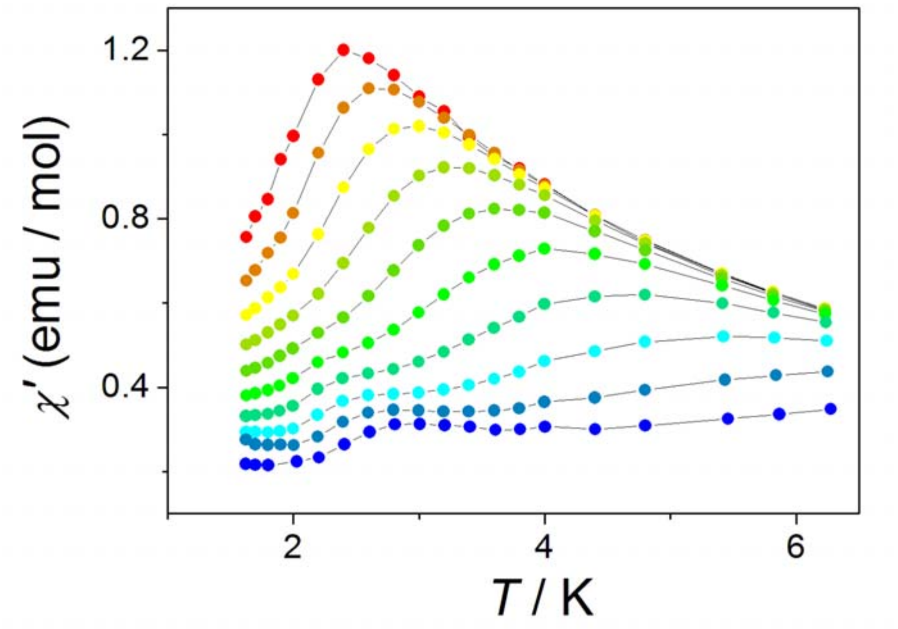

Fig. **S4**: Temperature dependence of χ' for a polycrystalline sample of the doped [Dy-Y NITpPy]$_2$ species, measured in 0.2 T static field from 1.6 to 6.2 K using ten logarithmically spaced frequencies, ranging from 100 Hz to 25 kHz.

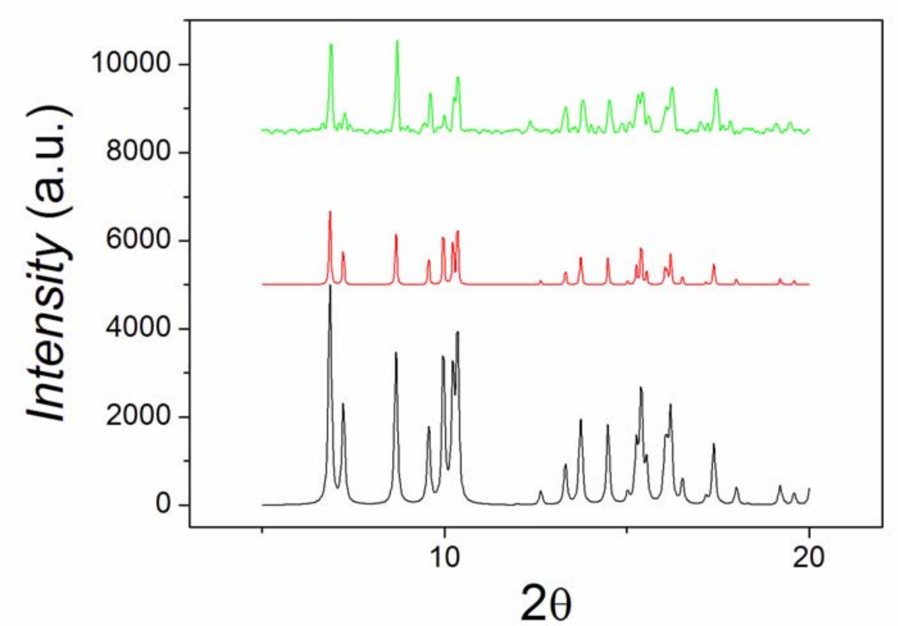

Fig. **S5**: Experimental powder diffraction patterns of [Dy-Y NITpPy]$_2$ (green, top), [DyNITpPy]$_2$ (red, middle) and simulated for [DyNITpPy]$_2$ (black, bottom). Experimental data were taken using a powder diffractometer Bruker D8 Avance working with Cu Kα radiation in *y/2y* mode. Theoretical signals were simulated from a CIF file with PowderCell software.

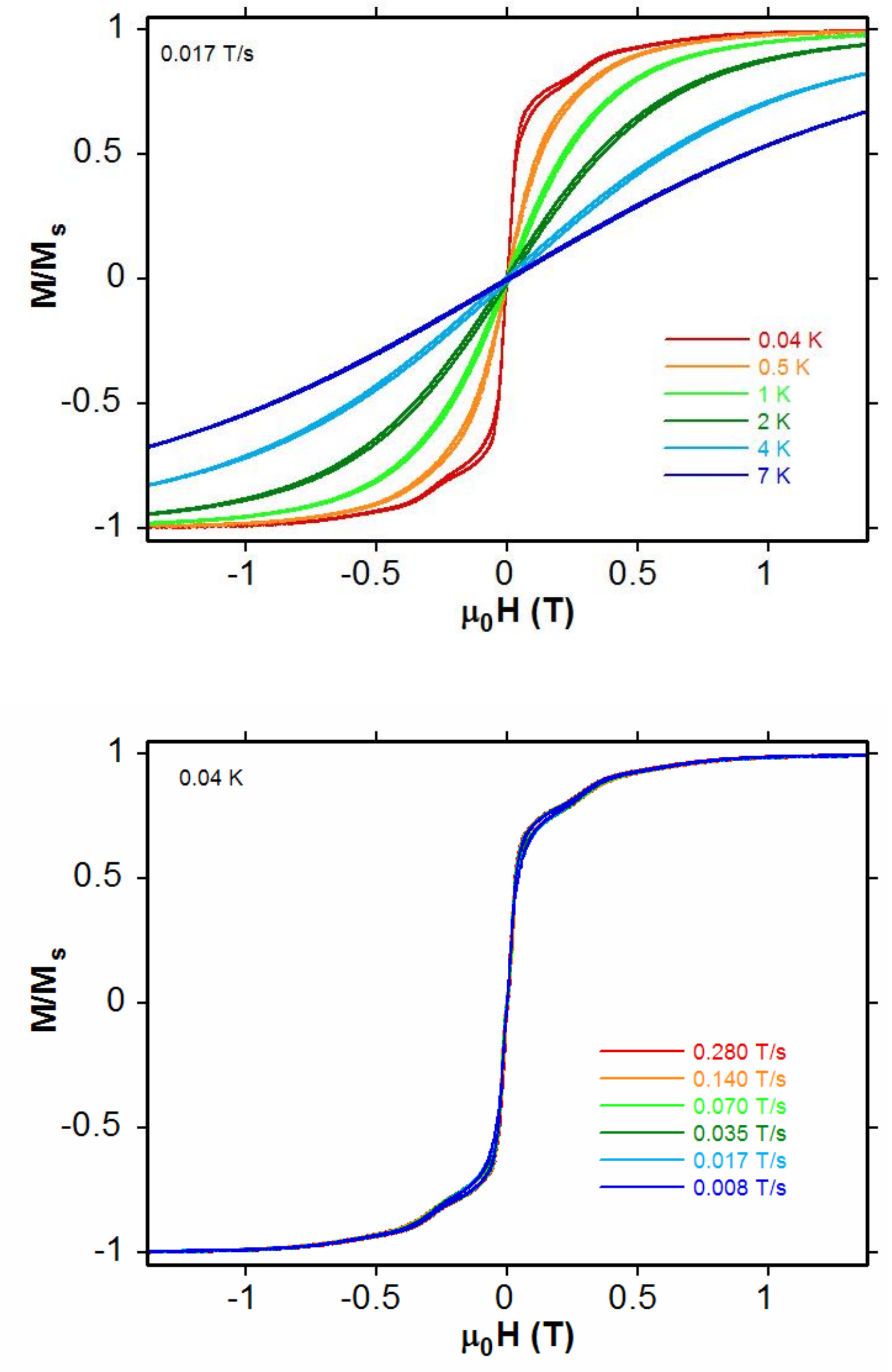

Fig. **S6**: Magnetization *vs* field curves for a single crystal of $[DyNITpPy]_2$ oriented along the easy axis of magnetization, taken for six different temperatures (above) and for six different field sweeping rates (below) using an array of micro-SQUIDs.