Strong carbon cage influence on the single molecule magnetism in Dy-Sc nitride clusterfullerenes

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Supporting Information

LDI mass-spectra of the studied compounds

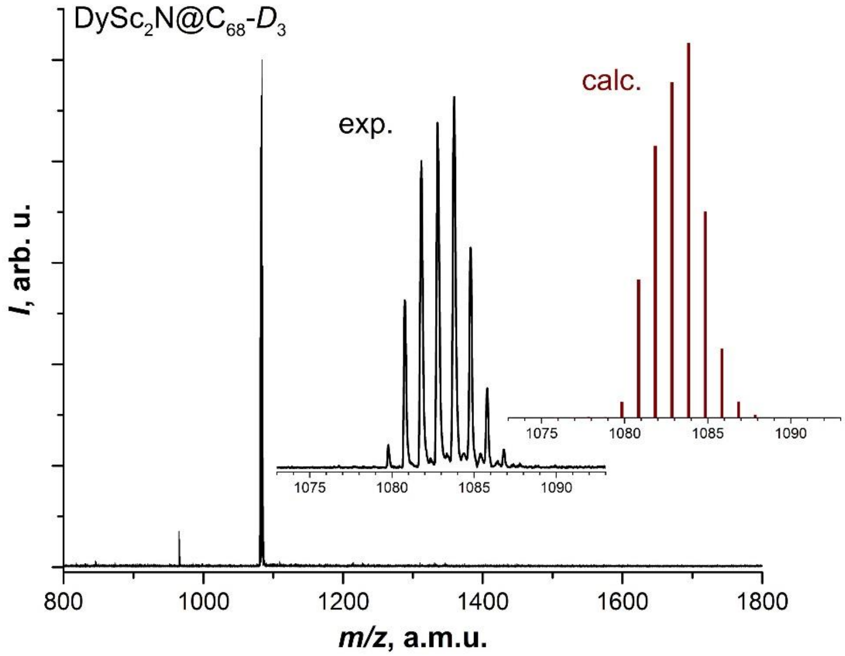


Figure S1. LDI Mass-spectrum of $DySc_2N@C_{68}$-D_3, positive ion mode. Insets show experimental and calculated isotope distribution.

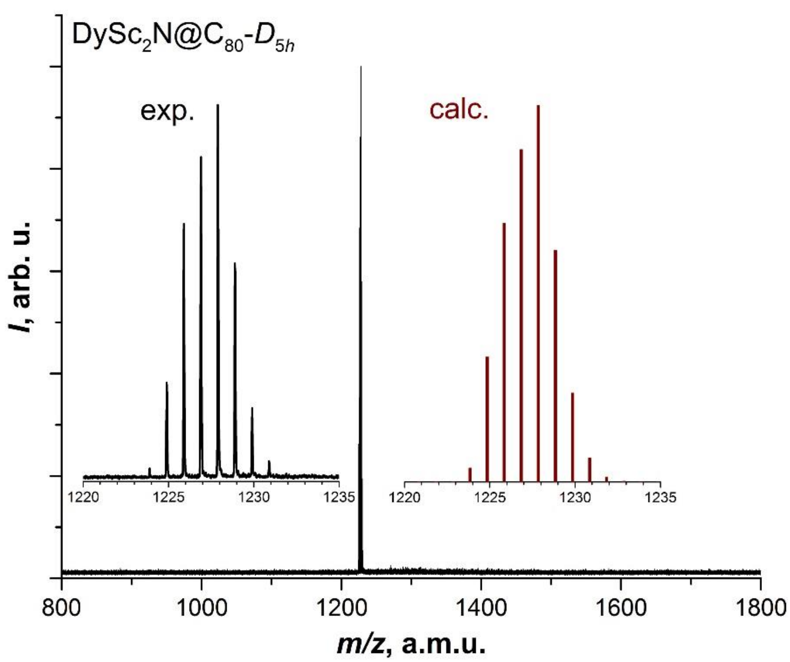


Figure S2. LDI Mass-spectrum of $DySc_2N@C_{80}$-D_{5h}, positive ion mode. Insets show experimental and calculated isotope distribution.

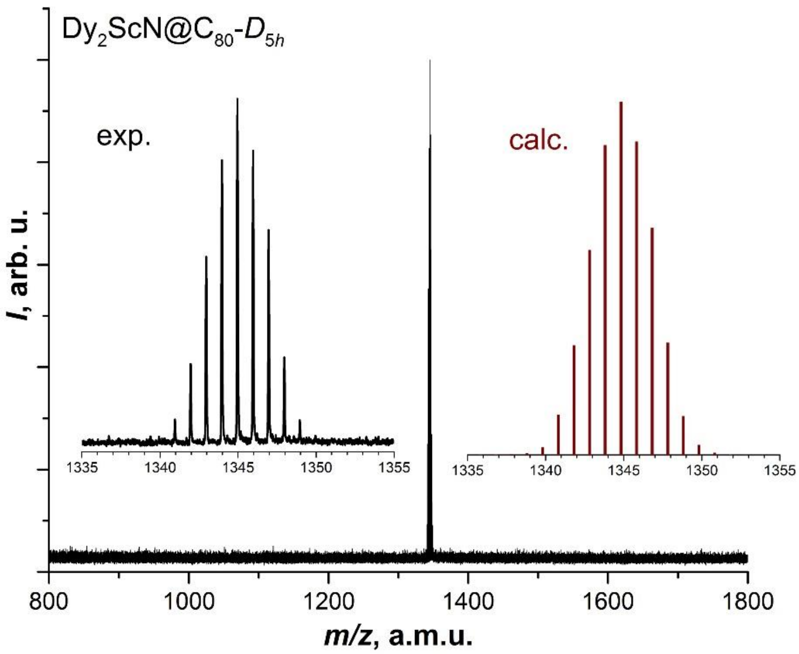


Figure S3. LDI Mass-spectrum of $Dy_2ScN@C_{80}$-D_{5h}, positive ion mode. Insets show experimental and calculated isotope distribution.

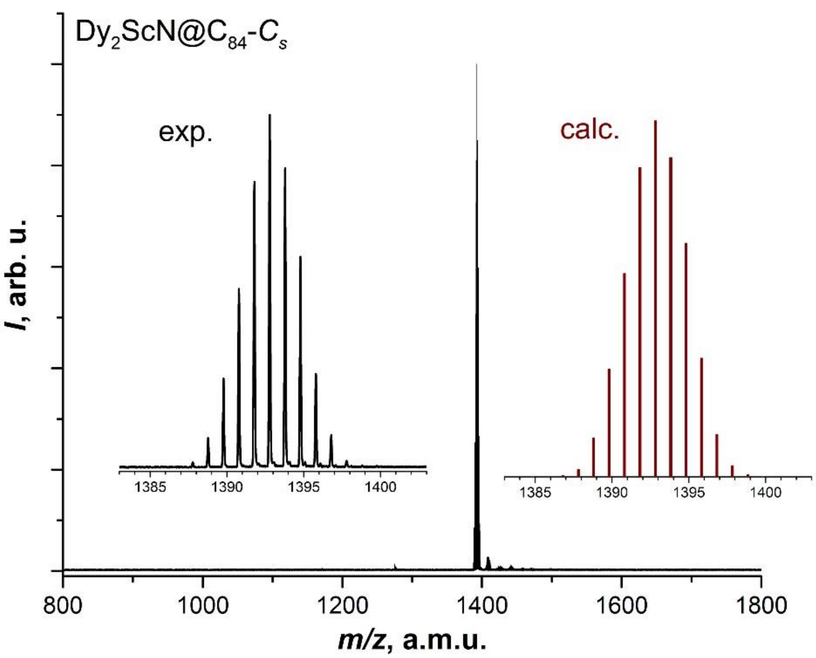


Figure S4. LDI Mass-spectrum of $Dy_2ScN@C_{84}$-C_s, positive ion mode. Insets show experimental and calculated isotope distribution.

Determination of relaxation times

The DC measurement of the magnetization decay curve is relatively simple and doesn't require large sample amounts (5-10 % compared to AC). However, difficulties arise during fitting of the experimental decay curves as single-molecule magnets tend to exhibit a time-dependent decay rate. One of the reasons is arising from the evolution of internal dipolar fields in the sample during relaxation. Consequently a single exponential function often fails to describe the system's behavior. In a general case the decay curve consists of an infinite number of exponentials, and characteristic value for the relaxation time distributions has to be derived. It becomes possible with a stretched exponential:

$$f(t) = M_{eq} + (M_0 - M_{eq})\, exp\left[-\left(\frac{t}{\tau_1}\right)^{\beta}\right] \quad \text{(S.1)}$$

Where M_{eq} and M_0 are the equilibrium and initial magnetizations, respectively, τ_1 is a characteristic "average" relaxation time and β is an additional parameter that corresponds to the time-dependent decay rate $\tau^{-1} \sim t^{\beta-1}$ with $\beta = (0; 1)$. In the extreme case of $\beta = 1$ one obtains a single exponential.

Relaxation of magnetization in $DySc_2N@C_{68}$-D_3

Table S1. Relaxation times and β-parameters from stretched exponential fitting of magnetization decays curves measured for $DySc_2N@C_{68}$-D_3 in a field of 0.2 T.

T, K	*τ*, s	st. dev. *τ*	β	st. dev. β
1.80	268.6	0.7	0.560	0.0007
2.00	195.8	0.7	0.560	0.0010
2.18	134.6	0.7	0.541	0.0013
2.35	97.4	0.7	0.590	0.0023
2.86	54.5	0.5	0.619	0.0039
3.18	40.3	0.9	0.492	0.0049

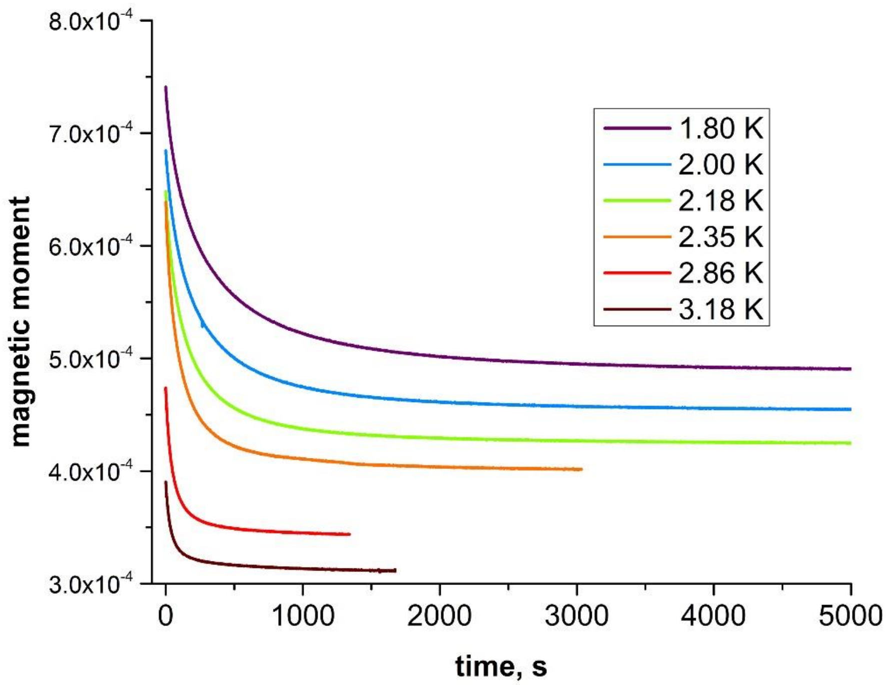


Figure S5. Magnetization decay curves measured for $DySc_2N@C_{68}$-D_3 at different temperatures in a field of 0.2 T

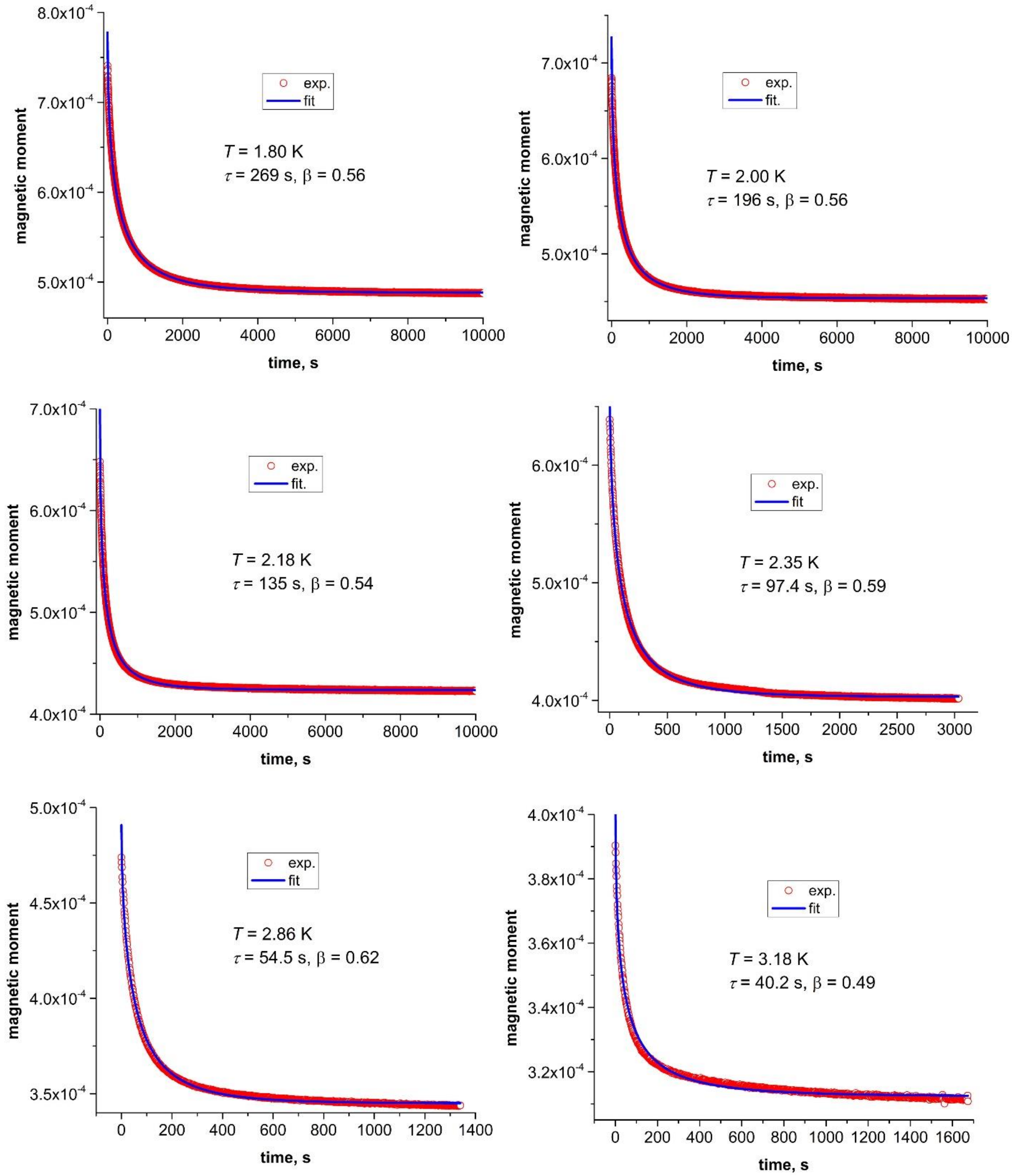


Figure S6. Fitting of experimental decay curves with stretched exponentials for $DySc_2N@C_{68}$-D_3

Relaxation of magnetization in $DySc_2N@C_{60}$-$\boldsymbol{D_{5h}}$

Table S2. Relaxation times and β-parameters from stretched exponential fitting of magnetization decays curves measured for $DySc_2N@C_{80}$-D_{5h} in a field of 0.2 T.

T, K	*τ*, s	st. dev. *τ*	β	st. dev. β
1.80	5234.5	5.7	0.719	0.0005
2.00	3228.7	3.2	0.693	0.0006
2.18	2014.4	1.9	0.685	0.0007
2.35	1313.7	1.3	0.667	0.0009
2.86	398.2	1.0	0.609	0.0012
3.18	207.4	0.7	0.633	0.0018
3.64	95.6	0.5	0.635	0.0024
4.00	55.4	0.5	0.622	0.0032

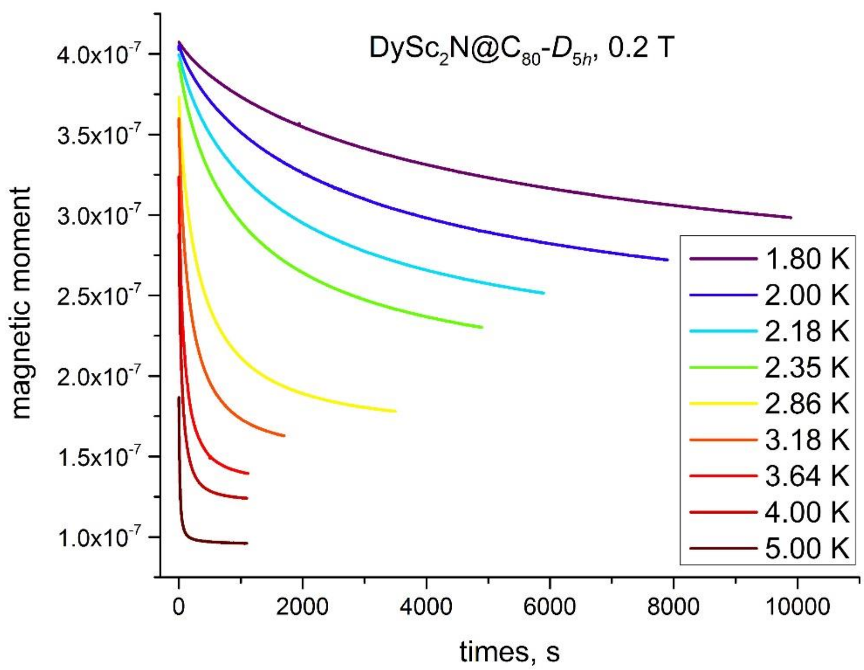
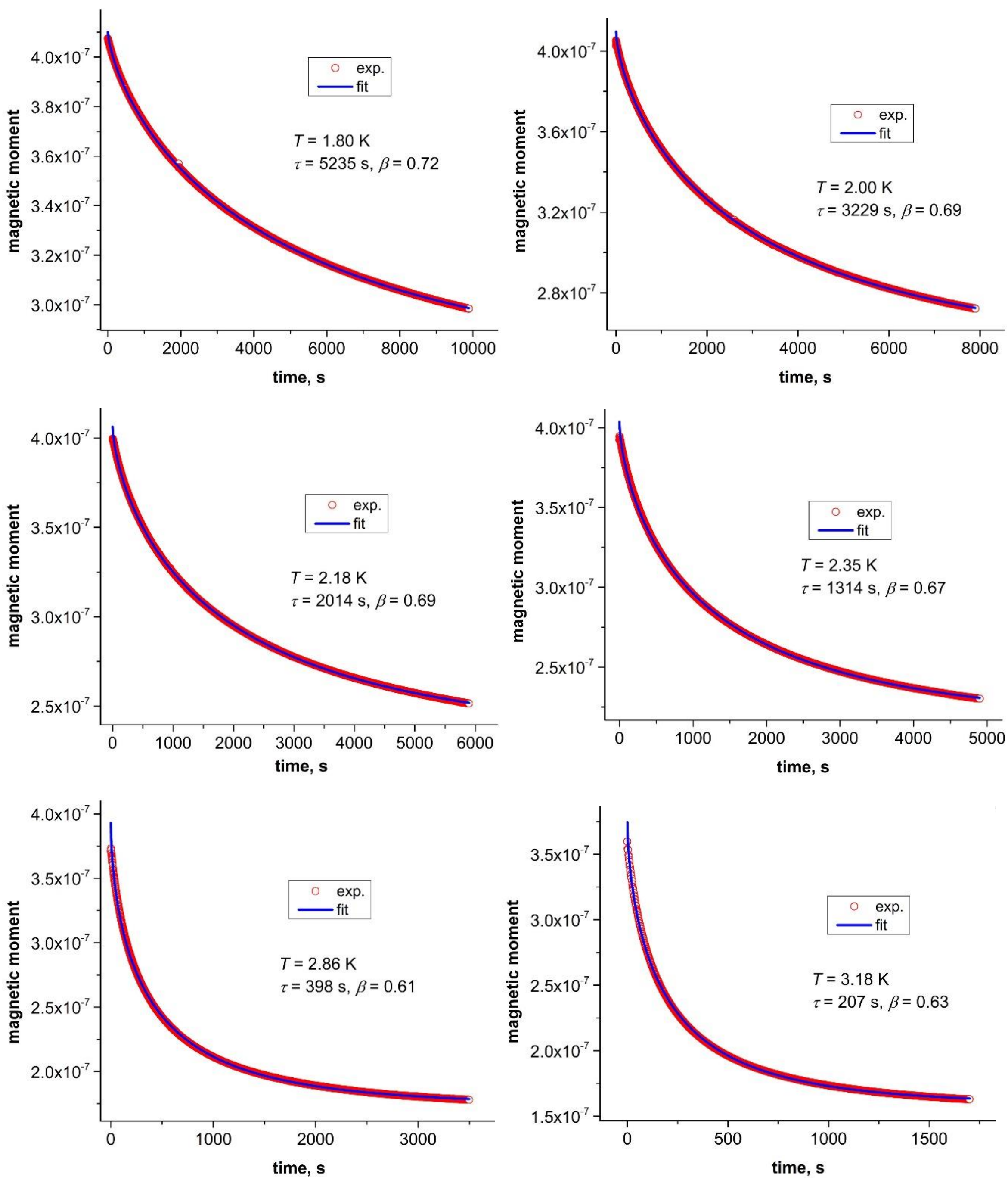


Figure S7. Magnetization decay curves measured for $DySc_2N@C_{80}$-D_{5h} at different temperatures in a field of 0.2 T



magnetic moment
time, s
exp.
fit
T = 1.80 K
τ = 5235 s, β = 0.72
4.0x10-7
3.8x10-7
3.6x10-7
3.4x10-7
3.2x10-7
3.0x10-7
0
2000
4000
6000
8000
10000
T = 2.00 K
τ = 3229 s, β = 0.69
4.0x10-7
3.6x10-7
3.2x10-7
2.8x10-7
T = 2.18 K
τ = 2014 s, β = 0.69
3.5x10-7
3.0x10-7
2.5x10-7
1000
3000
5000
T = 2.35 K
τ = 1314 s, β = 0.67
T = 2.86 K
τ = 398 s, β = 0.61
2.0x10-7
T = 3.18 K
τ = 207 s, β = 0.63
1.5x10-7
500
1500

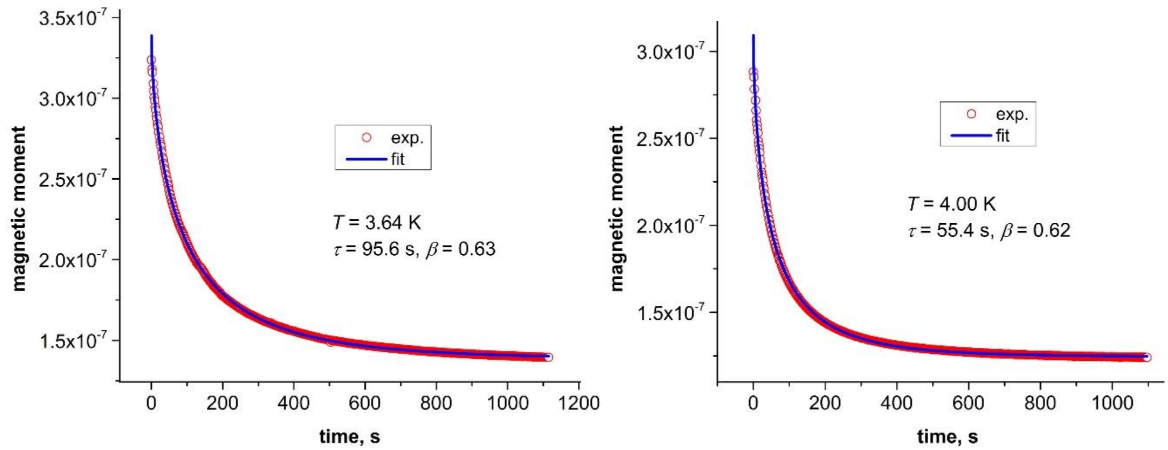


Figure S8. Fitting of experimental decay curves with stretched exponentials for $DySc_2N@C_{80}$-D_{5h}

Relaxation of magnetization in $Dy_2ScN@C_{80}$-D_{5h}

Table S3. Relaxation times and β-parameters from stretched exponential fitting of magnetization decays curves measured for $Dy_2ScN@C_{80}$-D_{5h} in zero field.

T, K	τ, s	st. dev. τ	β	st. dev. β
1.80	432.72	0.30	0.687	0.0003
2.00	286.91	0.26	0.701	0.0004
2.18	199.11	0.21	0.697	0.0005
2.35	147.10	0.17	0.695	0.0005
2.86	83.28	0.14	0.731	0.0009
3.18	54.32	0.12	0.726	0.0011
3.64	38.59	0.11	0.760	0.0016
4.00	31.17	0.14	0.782	0.0028

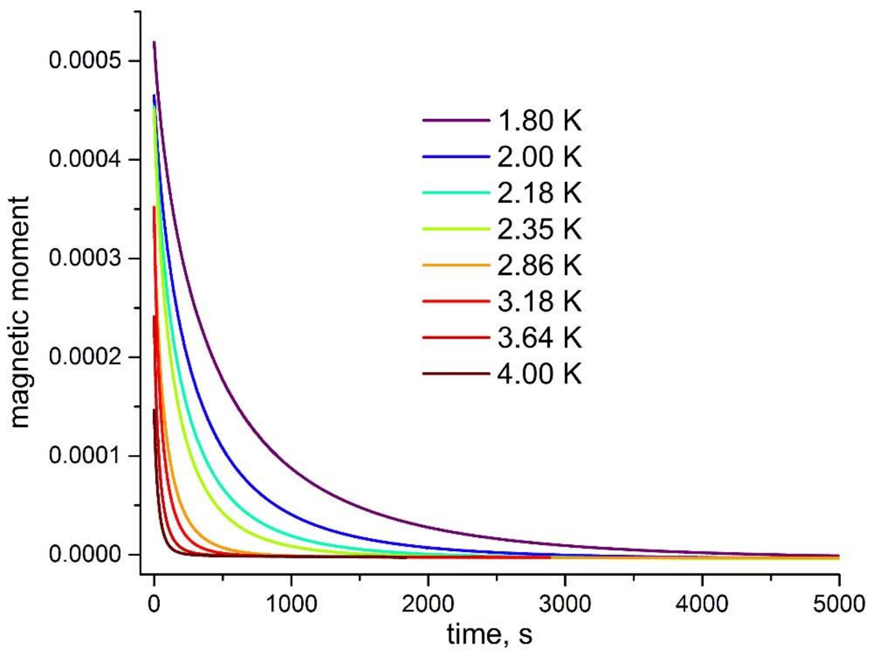
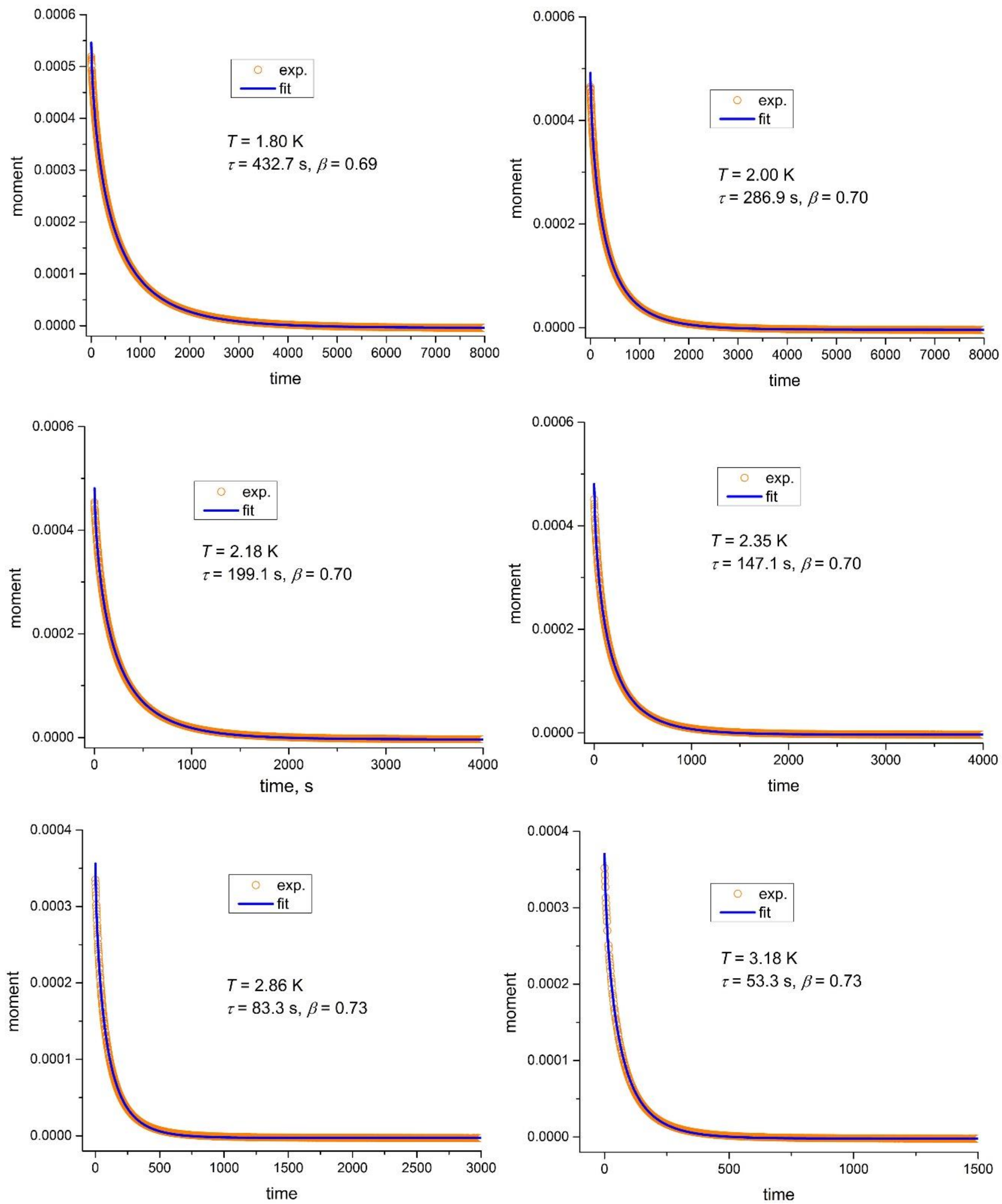


Figure S9. Magnetization decay curves measured for $Dy_2ScN@C_{80}$-D_{5h} at different temperatures



exp.
fit
T = 1.80 K
τ = 432.7 s, β = 0.69
moment
time
T = 2.00 K
τ = 286.9 s, β = 0.70
moment
time
T = 2.18 K
τ = 199.1 s, β = 0.70
moment
time, s
T = 2.35 K
τ = 147.1 s, β = 0.70
moment
time
T = 2.86 K
τ = 83.3 s, β = 0.73
moment
time
T = 3.18 K
τ = 53.3 s, β = 0.73
moment
time

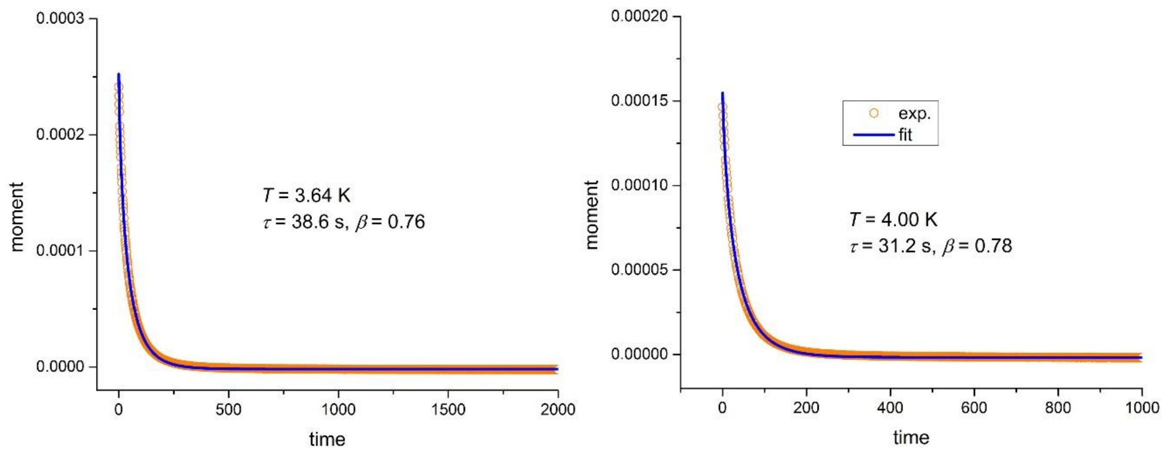


Figure S10. Fitting of experimental decay curves with stretched exponentials for $Dy_2ScN@C_{80}$-D_{5h}