

Figure S1 A packing diagram of **1** (hydrogen atoms omitted for clarity) showing ring interaction at ca 4 Å.

	low temperature, 90 K		room temperature, 296 K	
Formula	C31 H45 Co N3 O2		C31 H45 Co N3 O2	
<i>M</i>	550.63		550.63	
Crystal system	triclinic		triclinic	
space group	P-1		P-1	
<i>a</i> / Å	9.5337(6)		9.563(3)	
<i>b</i> / Å	11.7923(8)		12.063(1)	
<i>c</i> / Å	13.7991(10)		13.857(3)	
α / °	92.836(3)		93.60(1)	
β / °	109.513(3)		109.05(2)	
γ / °	99.029(3)		99.36(1)	
<i>V</i> /Å ³	1435.46(17)		1479.3(6)	
<i>Z</i>	2		2	
μ /mm ⁻¹	0.630		0.607	
λ	0.71073		0.71069	
Reflections	5040		4118	
collected/unique	3815		3367	
	refine_ls_R_factor_all	0.0574	refine_ls_R_factor_all	0.0486
	refine_ls_R_factor_gt	0.0357	refine_ls_R_factor_obs	0.0341
	refine_ls_wR_factor_ref	0.0843	refine_ls_wR_factor_all	0.0979
	refine_ls_wR_factor_gt	0.0772	refine_ls_wR_factor_obs	0.0895
Distances within	O1 Co1 1.9527(15)		Co1 O1 1.944(2)	
the chromophore	O2 Co1 1.9580(15)		Co1 O2 1.952(2)	
(different atom	N1 Co1 2.0516(18)		Co1 N1 2.045(2)	
labeling for N1 and	N2 Co1 2.1106(19)		Co1 N2 2.123(2)	
N3)	N3 Co1 2.0578(18)		Co1 N3 2.054(2)	

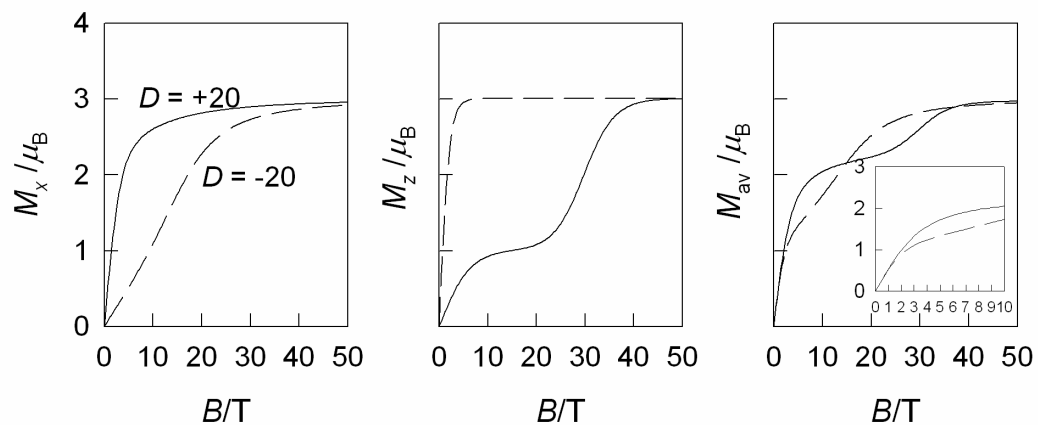


Figure S2 Modeling the components of the magnetization and its powder average using the exact partition function for the $S = 3/2$ system. Solid – positive D , dashed – negative D (in units of cm^{-1}); $T = 4.2$ K and $g = 2$. Saturation is not reached until $B = 10$ T.

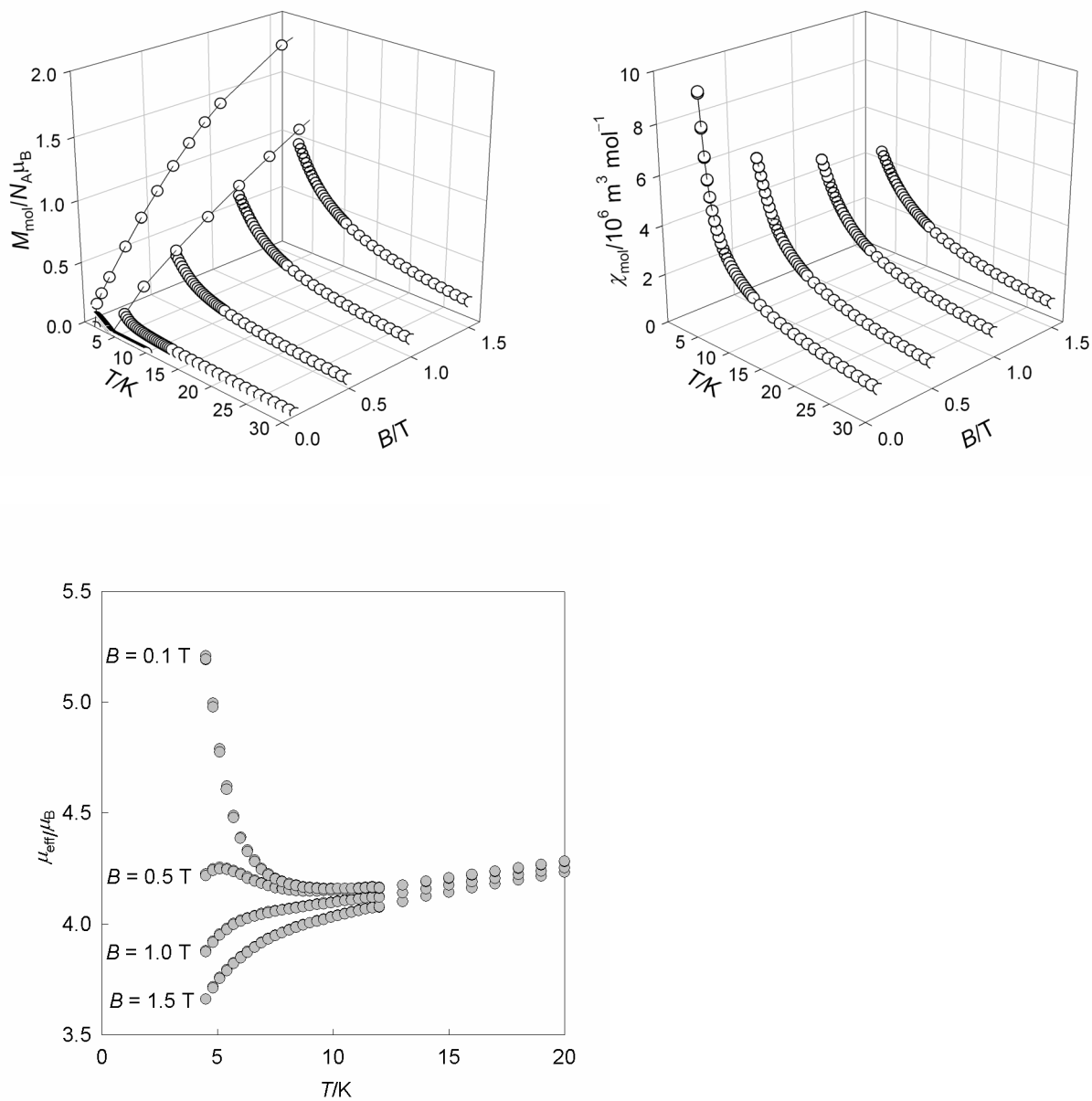


Figure S3 Temperature-field dependence of magnetization (top-left), mean magnetic susceptibility based upon magnetization (top-right) and effective magnetic moment (bottom-left) for **1**.

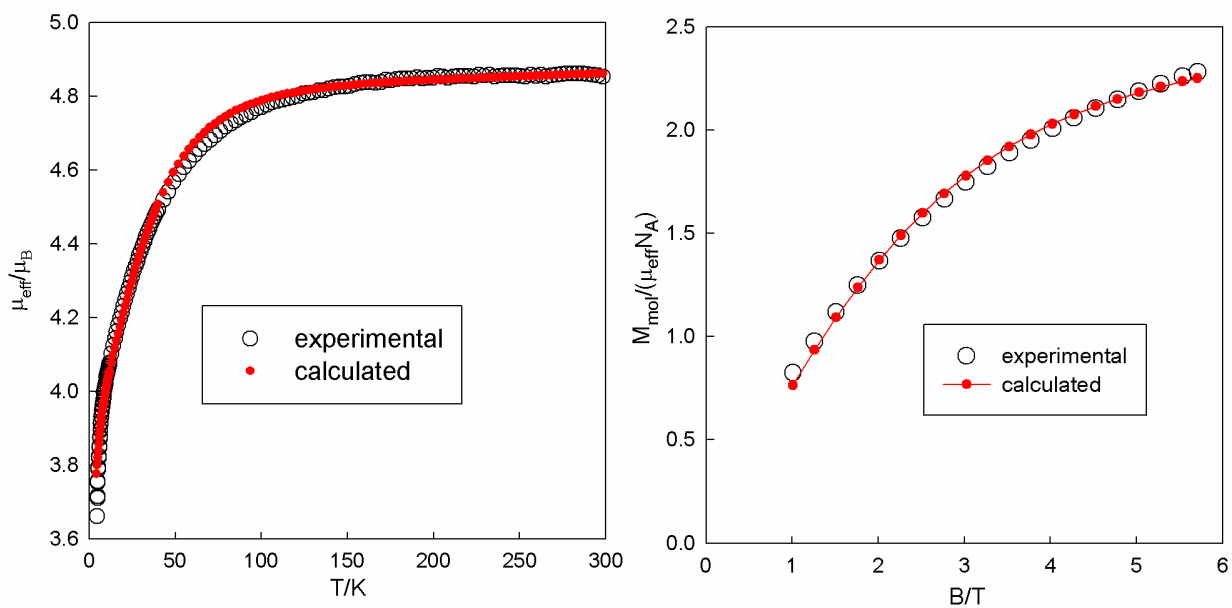


Figure S4 A simultaneous fit of the magnetic functions: Effective magnetic moment (SQUID, $B = 1.5$ T) – right. Magnetization at $T = 4.38$ K (left). Empty circles – experimental data; solid lines – calculated data.