Supplementary information for "A call for frugal modelling: two case studies involving molecular spin dynamics"

Gerliz M. Guti´errez-Finol,[1] Aman Ullah,[1] Mar´ıa González-B´ejar,[1] and Alejandro Gaita-Ariño[1,*]
[1]*Instituto de Ciencia Molecular (ICMol), Universitat de Val`encia, Paterna, Spain*
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CONTENTS

* alejandro.gaita@uv.es

S1. SPIN ENERGY LEVELS, VIBRONIC COUPLINGS AND SPIN-ELECTRIC COUPLINGS

Supplementary Table 1. Energy level scheme (in cm^{-1}) and predominant M_J microstate of the ground multiplet of the HoW_{10} complex calculated at CASSCF-SO and REC model implmented in SIMPRE on the crystalline coordinates.

$E^{(crys)}$(cm^{-1}), SIMPRE	M_J	$E^{(crys.)}$(cm^{-1}), CASSCF-SO	M_J
0.00	50 %$\|\pm 4\rangle$	0.00	47.5%$\|\pm 4\rangle$
0.32	50 %$\|\pm 4\rangle$	0.36	47.5%$\|\pm 4\rangle$
32.39	50 %$\|\pm 5\rangle$	26.24	47.2%$\|\pm 3\rangle$
32.76	50 %$\|\pm 5\rangle$	27.92	47.2%$\|\pm 3\rangle$
50.27	50 %$\|\pm 3\rangle$	50.08	48.7%$\|\pm 5\rangle$
53.33	50 %$\|\pm 3\rangle$	50.31	48.7%$\|\pm 5\rangle$
132.97	40 %$\|\pm 2\rangle$	86.70	48.2%$\|\pm 2\rangle$
138.95	36 %$\|\pm 2\rangle$	96.34	48.2%$\|\pm 2\rangle$
167.48	40 %$\|\pm 6\rangle$	155.59	48.9%$\|\pm 1\rangle$
169.32	36 %$\|\pm 6\rangle$	156.90	48.9%$\|\pm 1\rangle$
222.14	50 %$\|\pm 1\rangle$	178.90	46.6%$\|\pm 6\rangle$
226.39	50 %$\|\pm 1\rangle$	179.05	46.6%$\|\pm 6\rangle$
255.06	100 %$\|0\rangle$	181.88	94.3%$\|0\rangle$
330.90	50 %$\|\pm 7\rangle$	279.97	49.3%$\|\pm 8\rangle$
330.92	50 %$\|\pm 7\rangle$	279.97	49.3%$\|\pm 8\rangle$
379.51	50 %$\|\pm 8\rangle$	315.99	49.1%$\|\pm 7\rangle$
379.51	50 %$\|\pm 8\rangle$	315.99	49.1%$\|\pm 7\rangle$

Supplementary Table 2: Vibrational frequencies for normal mode-i, re- lated vibronic couplings (S_i) couplings calculated at REC model and CASSCF-SO method.

Mode (i)	Frequency (cm^{-1})	REC, S_i (cm^{-1})	CASSCF-SO, S_i (cm^{-1})
1	8.6188	0.0523	0.0848
2	17.9408	0.1352	0.1758
3	18.4270	0.1111	0.1190
4	24.3339	0.2348	0.2469
5	29.9667	0.2281	0.2196
6	43.2222	0.1668	0.1228
7	54.0211	0.1525	0.1502
8	58.3858	0.1867	0.1805
9	67.1204	0.1661	0.1454

10	70.9458	0.1423	0.1302
11	99.6054	0.0875	0.1249
12	106.5181	0.1242	0.1507
13	106.7797	0.0501	0.0508
14	107.2554	0.1062	0.1324
15	108.7554	0.1450	0.1667
16	114.6772	0.1045	0.0668
17	114.7083	0.1123	0.0804
18	140.1098	0.1336	0.1086
19	140.7649	0.0413	0.0400
20	141.1907	0.0538	0.0377
21	142.1063	0.1621	0.1290
22	143.9817	0.1441	0.1234
23	144.7529	0.0490	0.0393
24	157.6000	0.0213	0.0201
25	158.1734	0.0319	0.0810
26	159.1449	0.0282	0.0960
27	159.8794	0.0313	0.0422
28	160.5556	0.0225	0.0389
29	161.3001	0.0416	0.0795
30	167.7580	0.1431	0.0940
31	167.7659	0.1320	0.0833
32	184.6204	0.1046	0.1220
33	184.7316	0.0963	0.1219
34	193.0226	0.0473	0.0380
35	202.1425	0.1907	0.0948
36	216.4012	0.0333	0.0270
37	216.8891	0.0820	0.0656
38	218.1191	0.0985	0.0877
39	218.1262	0.1115	0.0992
40	218.2157	0.1155	0.1262
41	218.2437	0.1523	0.1171
42	219.9598	0.0635	0.0567
43	219.9980	0.0649	0.0544
44	220.0699	0.1268	0.1147
45	220.4673	0.1444	0.1107
46	223.1783	0.0980	0.0788
47	224.4059	0.1153	0.0724
48	230.8547	0.0421	0.0341
49	231.2420	0.0412	0.0314
50	231.7489	0.1052	0.0364
51	232.1038	0.1107	0.0360

52	235.1329	0.0205	0.0185
53	235.3319	0.0389	0.0292
54	257.4902	0.1404	0.0967
55	257.4918	0.1396	0.1076
56	289.5509	0.0197	0.0304
57	291.0713	0.0285	0.0248
58	329.1853	0.1204	0.0905
59	329.2450	0.1356	0.1248
60	337.2649	0.1024	0.1014
61	337.3633	0.1104	0.1426
62	337.4015	0.1131	0.0993
63	337.7207	0.1586	0.1013
64	351.1875	0.1267	0.1010
65	354.9521	0.1761	0.1475
66	355.2078	0.1900	0.1478
67	359.0168	0.1225	0.0640
68	360.2627	0.0433	0.0322
69	363.4859	0.1685	0.0848
70	399.0291	0.0518	0.0412
71	399.7840	0.0758	0.0671
72	399.8049	0.0757	0.0527
73	404.1887	0.1398	0.0829
74	414.5457	0.1438	0.1413
75	414.9182	0.1357	0.1258
76	416.6709	0.1086	0.1160
77	416.8970	0.1163	0.1110
78	417.5313	0.1370	0.1311
79	417.6863	0.1292	0.1203
80	429.9499	0.0752	0.0905
81	431.1486	0.0855	0.0650
82	435.1931	0.0777	0.0826
83	436.3070	0.0353	0.0455
84	436.7230	0.0605	0.0784
85	437.9110	0.0585	0.0565
86	479.0483	0.0218	0.0839
87	479.3424	0.0178	0.0698
88	480.9600	0.0378	0.0374
89	481.2860	0.0330	0.0378
90	489.3413	0.0814	0.0554
91	489.3780	0.0718	0.0543
92	514.2890	0.0288	0.0279
93	515.0904	0.0367	0.0306

94	515.2489	0.0249	0.0286
95	516.2136	0.1167	0.0761
96	520.3149	0.0824	0.1126
97	521.0873	0.0770	0.1017
98	528.0707	0.0683	0.0860
99	529.2183	0.0732	0.0793
100	530.4589	0.0552	0.0689
101	531.8096	0.0742	0.0801
102	535.5858	0.0644	0.0906
103	536.2067	0.0574	0.0966
104	554.9824	0.0325	0.0197
105	555.0443	0.0285	0.0184
106	578.4330	0.0189	0.0313
107	579.0391	0.0232	0.0425
108	579.1319	0.0141	0.0268
109	579.4927	0.0260	0.0334

Supplementary Table 3: The shift in transition frequency (δf) (MHz) obtained at different level of computation with applied voltage V.

Applied Voltages (volts)	Experimental	REC (opt.)	REC (crys.)	CASSCF-SO (opt.)	CASSCF-SO (crys.)
0	0.0000	0.0000	0.0000	0.0000	0.0000
50	–	0.1548	1.3621	0.0514	0.1105
100	0.5079	0.3096	2.7243	0.0970	0.2221
150	0.8447	0.4644	4.0866	0.1481	0.3347
200	1.1401	0.6193	5.4490	0.2016	0.4483
250	1.4200	0.7742	6.8116	0.2499	0.5630
300	1.7039	0.9292	8.1743	0.3032	0.6788

S2. DETAILED INSTRUCTIONS FOR REPRODUCIBILITY: SECOND CASE OF STUDY

This part offers a help for reproducing the graphics shown at Guti´errez-Finol et al. [1], but using the new version of STOSS in Matlab. We used Matlab R2023b, 64-bit and there is no need to download other files apart from the following ones:

1. main.m
2. read_data.m (description of the system).
3. mag_relaxation.m (relaxation Mechanisms, Total probability for spin flipping).
4. solve_single_p.m (single probabilities to pass from 0 to 1, and vice versa).
5. total_probability.m (total probability).
6. Bolztmann_distribution.m (zeeman effect, Boltzmann distribution).
7. changeable_field.m (analysis of conditional for spin flipping).
8. iteration_process.m (analysis of conditional for spin flipping).
9. mean_matrix_state.m (for a two p-bit network, where the collective state is studied in this function).
10. td.m (counting coincidences of state in both p-bits applying a delayed in the response).
11. association.m (for a two p-bit network, the association factor is calculated).
12. plotting.m (graphical representation of the results).
13. results.py (python script for saving the results).
14. full_data_file.csv (file which contains all the information for few systems, from the SIMDAVIS dataset)
15. user_configurations.xlsx (file which contains all the information about the system, here the user specifies the parameters).

It is important to emphasize that the file named *main.m* contains the body of the simulator, the user must not change any part of the script. As the older version of STOSS, we are capable to simulate three main scenarios:

1. Magnetization decays at different temperatures at constant magnetic field.
2. Magnetization decays at different temperatures at changeable magnetic field.
3. Magnetization decays at different temperatures of two p-bit network.

Considering this idea, the user can select the type of simulation just writing the values in each variable as it is shown in Supplementary Figure 1.

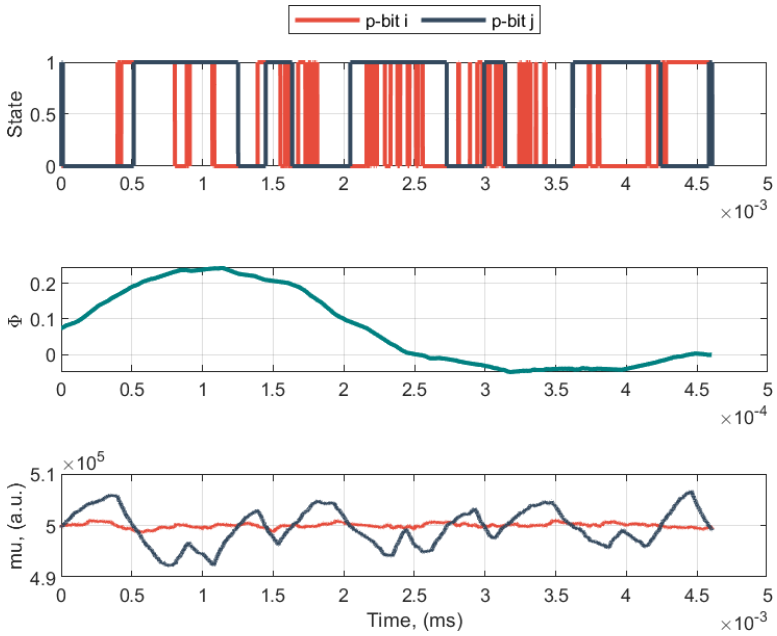
Given the explanation of the case of study in the body of the paper, this part follows the calculations of two p-bit network. To reproduce the result, we present the Supplementary Table 4 with all the necessary values and its simulation (Supplementary Figure 2). In contrast, Supplementary Figure 3 shows the summary part where we could see the computational cost when we increase the number of spins of each p-bit to 1 million with 10,000 total time steps using Matlab.

1		
2	**GENERAL CONFIGURATIONS**	
3		
4	**N_EX**	**1000000**
5	**T**	**4**
6	**save**	**0**
7	**flag**	**27**
8	**starting_mode**	**0.5**
9	**time_steps**	**10000**
10	**option**	**0**
11	**option_2spin**	**1**
12		
13	**CONSTANT MAGNETIC FIELD**	
14		
15	**B_constant**	**0**
16		
17	**CHANGEABLE MAGNETIC FIELD**	
18		
19	**B_max**	**0.00025**
20	**cycles**	**4**
21		
22	**TWO P-BITS NETWORK**	
23		
24	**B_pbit2**	**0.02**
25	**factor**	**9000**
26	**association_factor**	**1000**
27	**step_association_fact**	**1**
28		

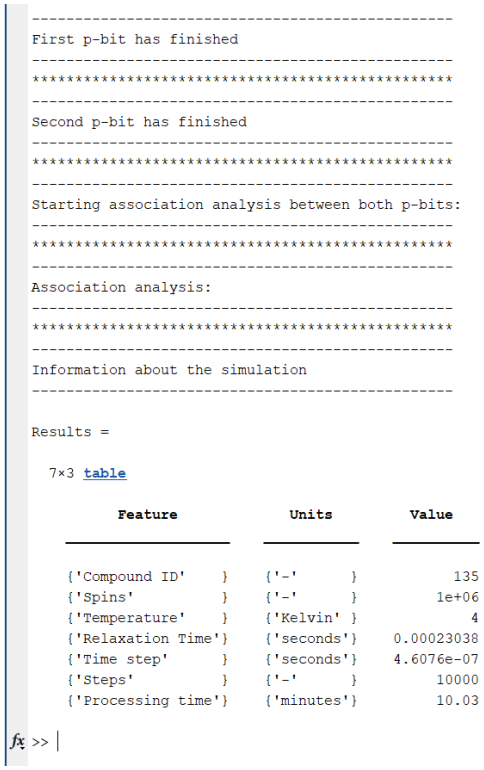
Supplementary Figure 1. Parameters that can be modified by the user (user configuration.xlsx).

Supplementary Table 4. Parameters at 4 K to analyze the effect of a p-bit to another, for [Dy(obPc)$_2$] Cd[Dy(obPc)$_2$].

Variable	Figure 2
N_ex	1000000
T	4
save	1
flag	25
starting mode	0.5
time steps	10000
option	0
option 2spin	1
B constant	0
B_max	DNA
cycles	DNA
B pbit2	0.02
factor	1000
association factor	time steps
step association factor	1



Supplementary Figure 2. Simulation employing N = 10^6 spins per p-bit at 4 K for [Dy(obPc)$_2$] Cd[Dy(obPc)$_2$]. Where part a) represents the state of each p-bit, b) the association factor, and c) the relaxation behaviour.



```
--------------------------------------------------
First p-bit has finished
--------------------------------------------------
**************************************************
--------------------------------------------------
Second p-bit has finished
--------------------------------------------------
**************************************************
--------------------------------------------------
Starting association analysis between both p-bits:
--------------------------------------------------
**************************************************
--------------------------------------------------
Association analysis:
--------------------------------------------------
**************************************************
--------------------------------------------------
Information about the simulation
--------------------------------------------------

Results =

  7×3 table

        Feature             Units          Value
    ________________    ___________    _________

    {'Compound ID'    }    {'-'      }          135
    {'Spins'          }    {'-'      }        1e+06
    {'Temperature'    }    {'Kelvin' }            4
    {'Relaxation Time'}    {'seconds'}    0.00023038
    {'Time step'      }    {'seconds'}    4.6076e-07
    {'Steps'          }    {'-'      }        10000
    {'Processing time'}    {'minutes'}        10.03

fx >>
```

Supplementary Figure 3. Simulation employing N = 10^6 spins per p-bit at 4 K for $[Dy(obPc)_2]$ $Cd[Dy(obPc)_2]$.

SUPPLEMENTARY REFERENCES

[1] Gutiérrez-Finol GM, Giménez-Santamarina S, Hu Z, Rosaleny LE, Cardona-Serra S, Gaita-Ariño A. Lanthanide molecular nanomagnets as probabilistic bits. npj Computational Materials. 2023;9(1):196.