Supporting Information

Computational Insights into the Encapsulation of a Dy(III) Bis(Amide)–Alkene Single-Ion Magnet within Porous Frameworks

Garima Bangar and Gopalan Rajaraman*

Department of Chemistry, Indian Institute of Technology Bombay, Powai, Mumbai 400076, India. Tel: (+91)-22-2576-7183. Email: rajaraman@chem.iitb.ac.in

Computational Details

Periodic DFT (pDFT) calculations were performed using CP2K[1] and VASP[2] within the PBE exchange–correlation functional including D3 dispersion correction (Becke–Johnson damping).[3, 4] In CP2K, the GAPW formalism was employed with DZVP-MOLOPT-GTH basis sets and GTH pseudopotentials, using a plane-wave cutoff of 400 Ry. In VASP, the PAW method with a plane-wave cutoff of 520 eV was employed, along with an electronic convergence criterion of 10^{-5} eV and a force convergence threshold of 0.02 eV $Å^{-1}$. For computational efficiency and to avoid complications arising from 4f electrons in periodic calculations, the Dy^{3+} centres were replaced by isostructural Y^{3+} ions in the VASP simulations.

All the ab initio calculations have been performed by the post-Hartree-Fock–Fock *ab initio* calculations by using the MOLCAS 8.2 code.[5] Basis sets for all atoms were taken from the ANO-RCC library as implemented in the MOLCAS package (Table **S1**).

Table S1. List of the used ANO-RCC basis sets.

Element	**Basis Sets**
Dy	ANO-RCC...8s7p5d3f2g1h.
C	ANO-RCC...3s2p1d
H	ANO-RCC...2s1p.
Si	ANO-RCC...4s3p1d.
N	ANO-RCC...3s2p1d.
Cl	ANO-RCC...4s3p1d.

All the calculations were carried out using complete active space (CAS). In the case of Dy, the active space comprised nine active Dy^{III} electrons in the seven 4f active orbitals RAS (9,7). The ground-state f-electron configuration for Dy^{III} is found to be $4f^9$, which yields a $^6H_{15/2}$ multiplet as the ground state. The CASSCF calculations were performed with the same basis sets and active spaces to obtain relative energies of the Kramers doublets (KDs), g tensors. For Dy^{III}, we used 21 roots in the sextet state in the configuration interaction (CI) procedure. Once these spin-free excited states are obtained, we mixed all of these states by using the RASSI-SO module to compute the spin-orbit coupled states. We obtained eight low-lying KDs for the Dy^{III} ion, which finally yielded g-tensors.

David P. Mills and co-workers originally synthesised complex **1**.[6] *Ab initio* CASSCF [7]calculations were first performed on both crystallographically resolved components of the X-ray structure (major and minor occupancies)(Figure **S1**, **S6a**, **S6b**), as well as on the fully optimised isolated cation (**1**$^+$), and two independently optimised models incorporating a proximal counteranion, namely $\mathbf{1}_{\mathbf{Cl}}$ and $\mathbf{1}_{\mathbf{Al(OCF3)4}}$, each considered separately to ensure charge neutrality (Figure **S2b/c**).

Structural Models and Optimised Geometries

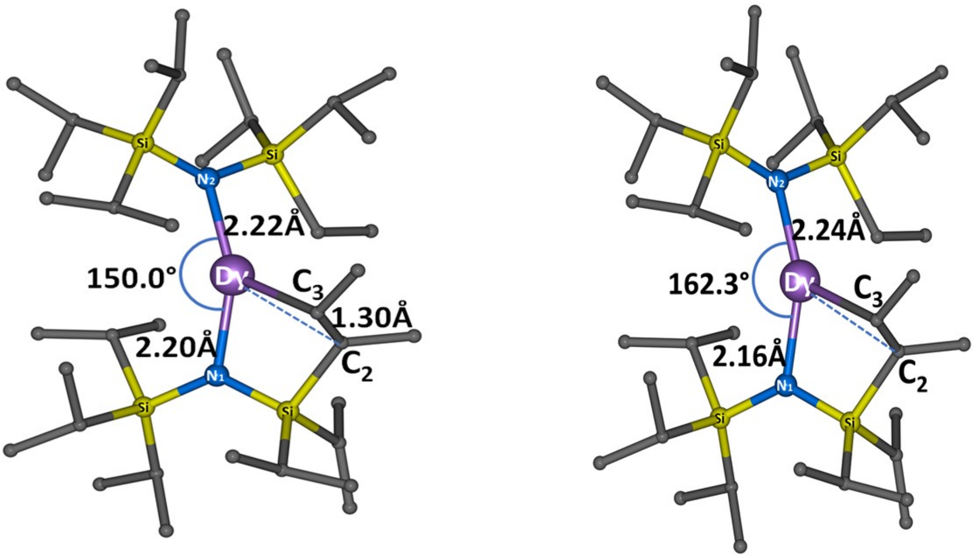

Figure S1: Crystal structure of 1_{xray}. The left side represents the major structure, and the right side is the minor structure according to the experiment.

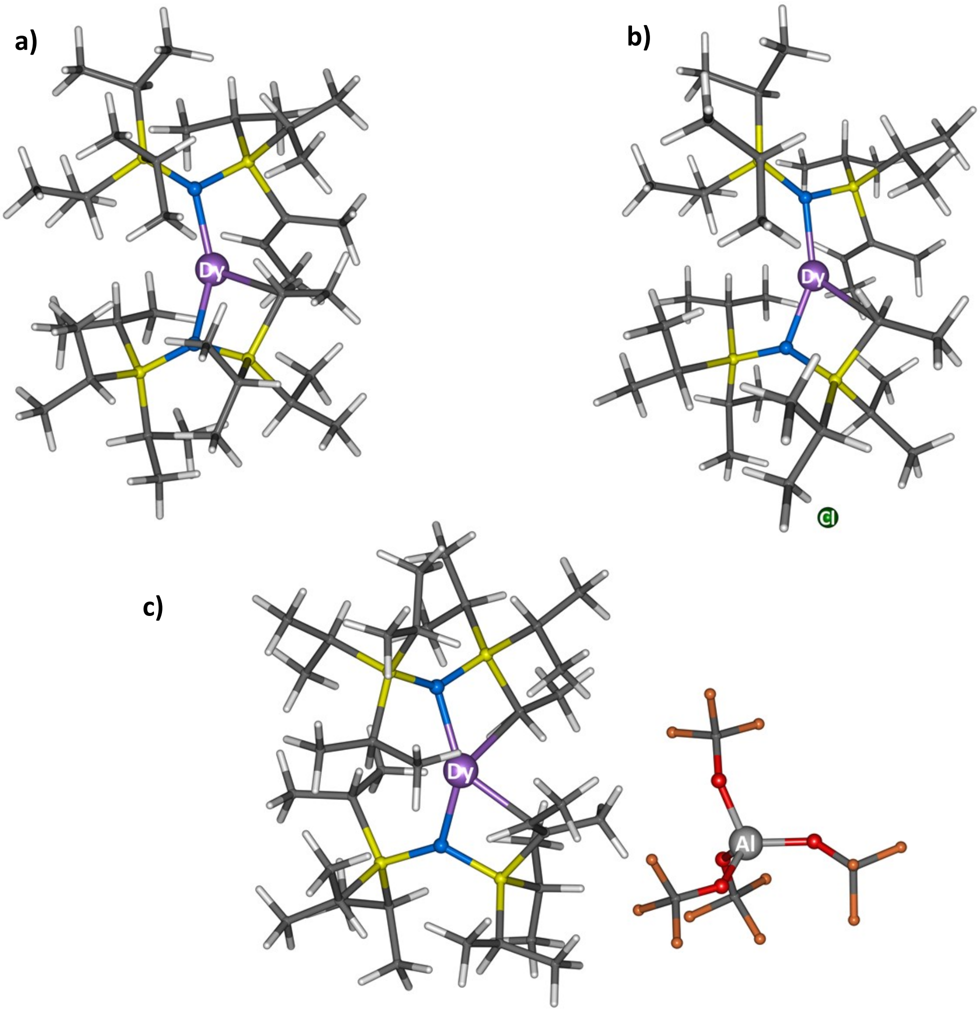

Figure S2: Optimised molecular structure of Dy complex a) $\mathbf{1}^+$, b) $\mathbf{1}_{\mathbf{Cl}}$ and c) $\mathbf{1}_{\mathbf{Al(OCF_3)_4}}$. Colour Code: Dy- Purple, N-Blue, Si-Yellow, C-Grey, Al-Light Grey, H-White, O-Red, F- Orange and Cl-Green.

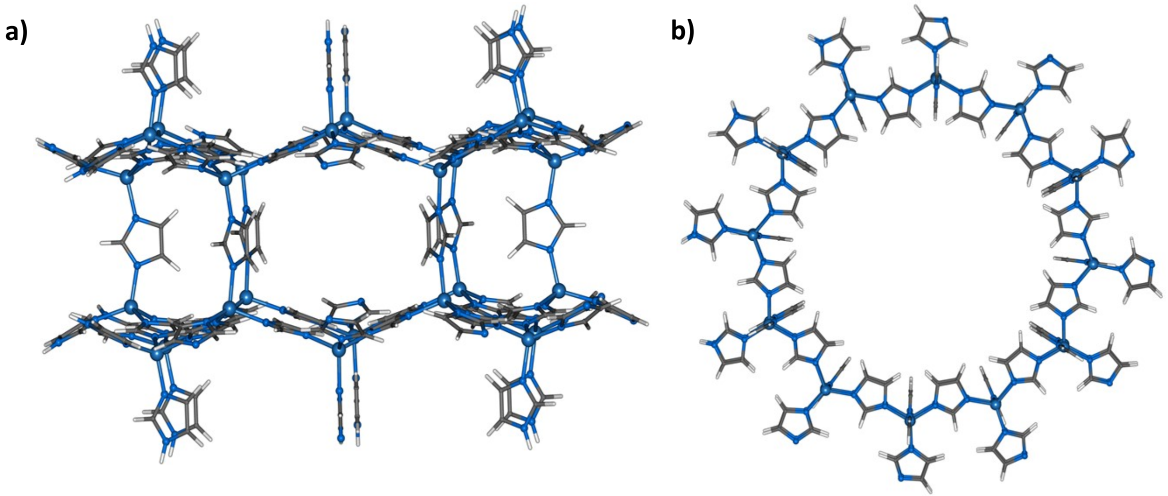

Figure S3: Unit cell of AFI-$[Zn(Im)_2]$ (**F1**) a) front view, and b) top view. Colour code: Zn: dark blue, N: Blue, C- grey and H- white.

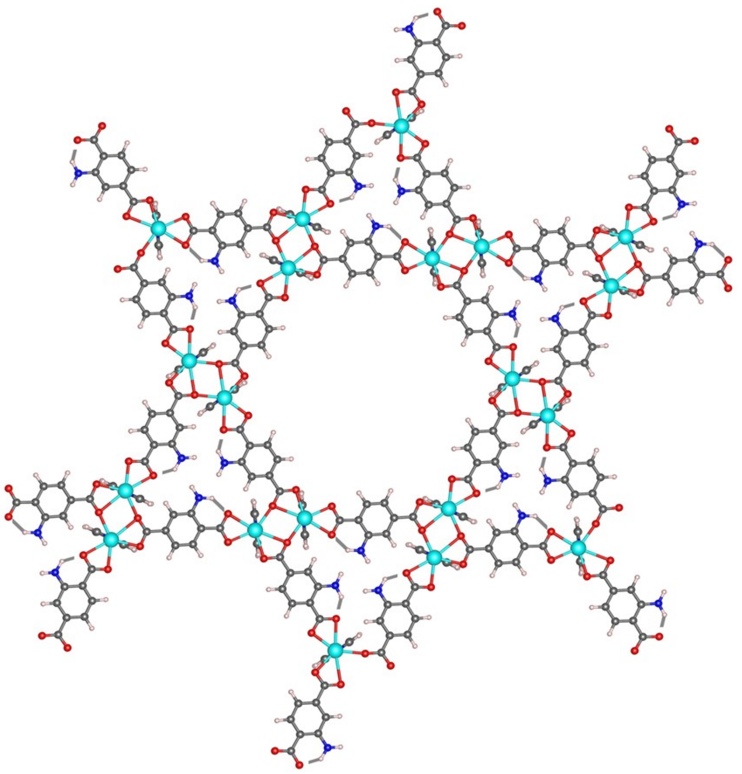

Figure S4: Unit cell of Cd-MOF (**F2**). Colour code: Cd-Cyan Blue, O-Red, N- Blue, C- grey, and H- white.

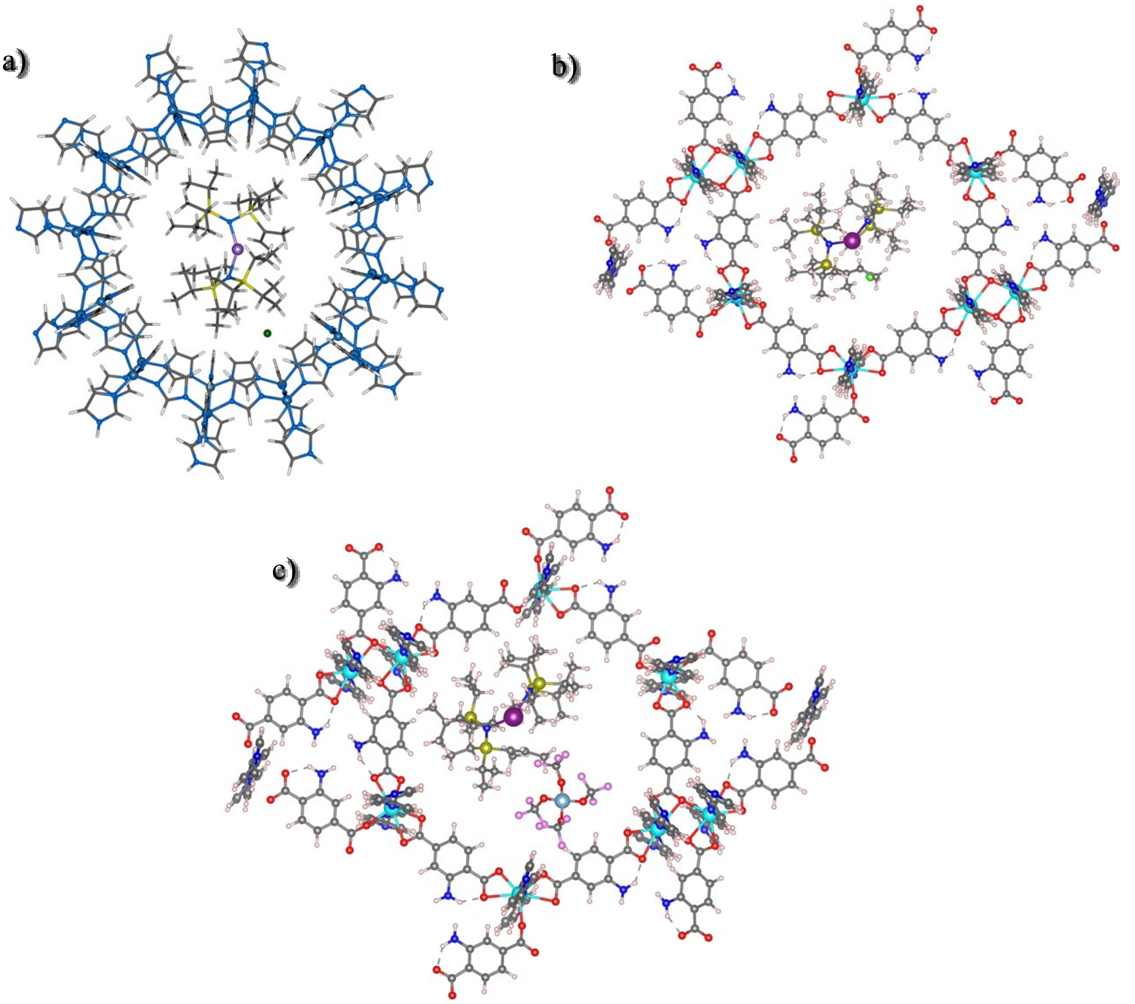

Figure **S5**: Optimised geometry of a) **1**$_{\mathrm{Cl}}$@**F1**, **1**$_{\mathrm{Cl}}$@**F2** and **1**$_{\mathrm{Al(OCF3)4}}$@**F2**.

Magnetic Anisotropy Data

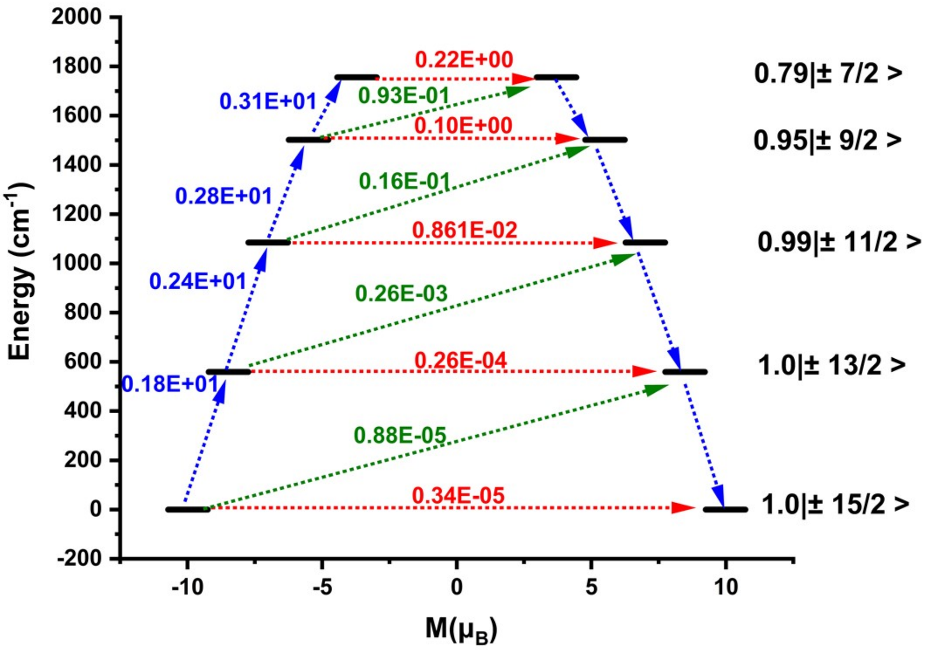

Figure S6a: *Ab initio* computed magnetisation blockade barriers for **1**$_{\textbf{x-ray}\text{(major)}}$

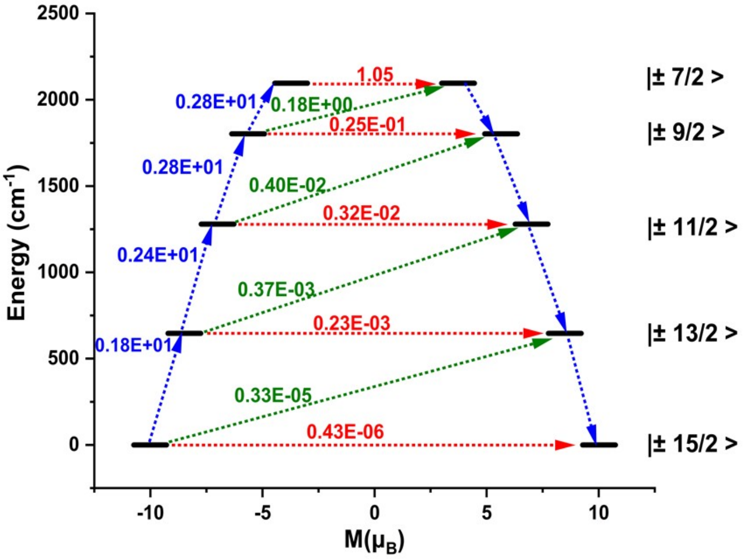

Figure S6b: *Ab initio* computed magnetisation blockade barriers for **1**$_{\textbf{x-ray}\text{(minor)}}$

Table S2. CASSCF+RASSI-SO computed energies of KDs and SINGLE_ANISO computed g-tensors of $\mathbf{1}_{\text{x-ray(major)}}$, $\mathbf{1}^+$, and $\mathbf{1}_{\text{Cl}}$

KDs	E(cm^{-1})	g_x	g_y	g_z	ϑ
$\mathbf{1}_{\text{xray}}$					
15/2	0.0	0.000	0.000	19.970	~
13/2	559.4	0.000	0.000	16.957	0.9
11/2	1084.7	0.024	0.027	14.013	1.6
9/2	1501.7	0.302	0.322	11.327	13.5
7/2	1755.8	0.256	0.757	9.642	39.8
5/2	1894.2	8.100	6.257	3.774	13.1
3/2	1992.7	1.137	3.241	14.645	88.0
1/2	2027.8	0.575	1.094	19.288	91.1
$\mathbf{1}^+$					
15/2	0.0	0.000	0.000	19.822	~
13/2	501.1	0.001	0.001	16.812	0.5
11/2	959.3	0.062	0.071	13.893	3.2
9/2	1315.7	0.631	0.651	11.212	15.3
7/2	1544.2	0.497	1.508	8.393	34.2
5/2	1676.6	3.991	5.936	8.647	94.1
3/2	1770.8	1.296	2.335	15.168	84.2
1/2	1790.8	0.621	1.191	19.205	94.4
$\mathbf{1}_{\text{Cl}}$					
15/2	0.0	0.000	0.000	20.003	~
13/2	553.8	0.002	0.002	17.024	2.1
11/2	1062.0	0.014	0.015	14.135	0.9
9/2	1461.1	0.275	0.317	11.484	7.9
7/2	1695.9	2.367	3.288	10.018	36.9
5/2	1796.4	0.381	3.291	12.682	69.5
3/2	1861.3	0.101	1.985	6.599	70.2
1/2	1920.2	12.469	8.624	1.232	16.6
$1_{\text{Al(OCF3)4}}$					
15/2	0.0	0.000	0.000	19.914	~
13/2	475.2	0.002	0.002	16.912	0.6
11/2	903.9	0.072	0.085	13.963	4.7
9/2	1237.8	0.413	0.450	11.202	16.5
7/2	1462.9	1.310	2.307	8.470	32.0
5/2	1600.8	8.657	7.209	3.626	2.5
3/2	1725.8	0.812	1.388	16.206	85.1
1/2	1781.6	0.136	0.254	19.630	95.4

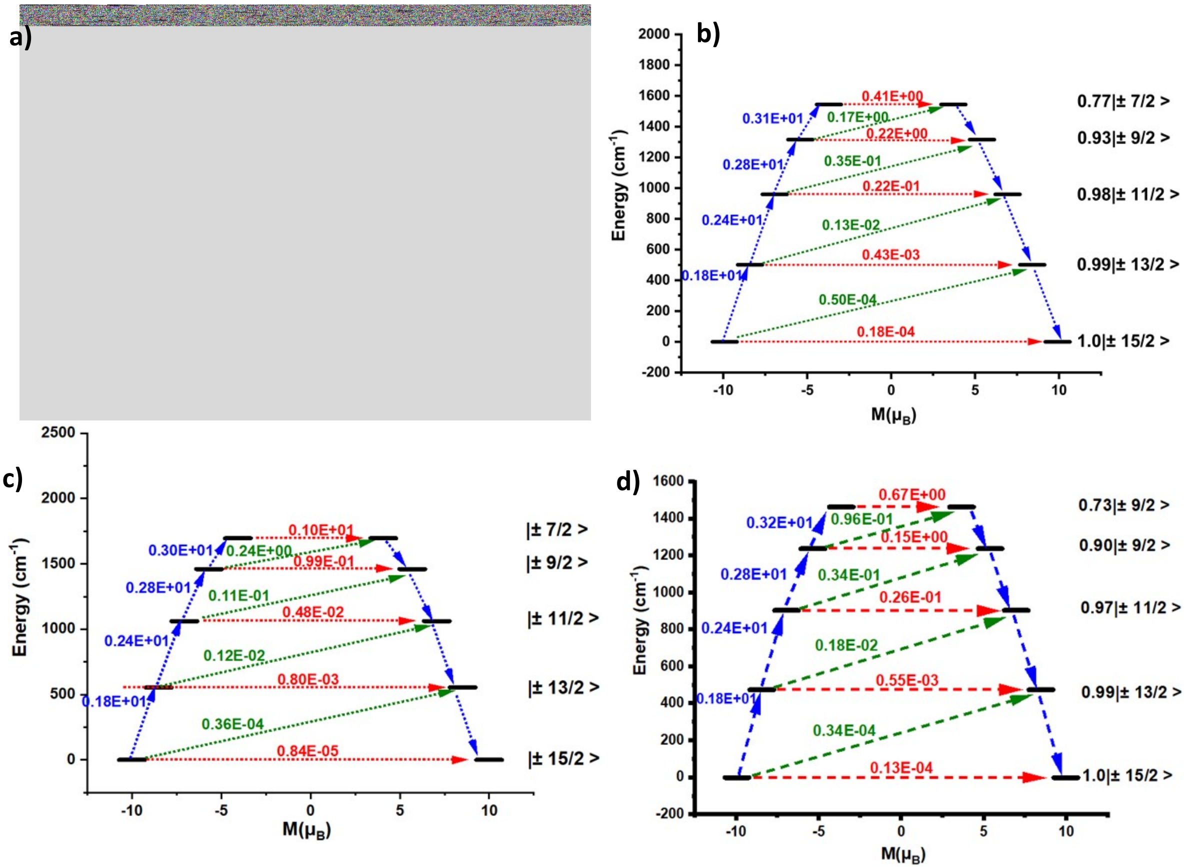

Figure S7: *Ab initio* computed magnetisation blockade barriers for a) $\mathbf{1}_{\text{x-ray}}$, b) $\mathbf{1}^{+}$, c) $\mathbf{1}_{\text{Cl}}$, and $\mathbf{1}_{\text{Al(OCF3)4}}$

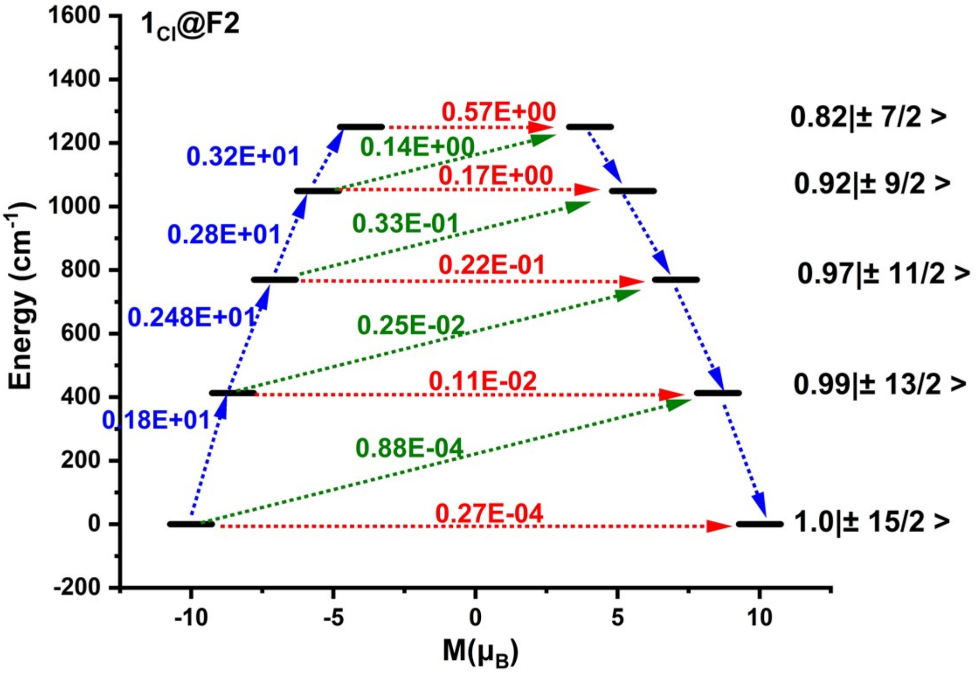

Figure S8: *Ab initio* computed magnetisation blockade barriers for $\mathbf{1}_{\text{Cl}}$**@F2**.

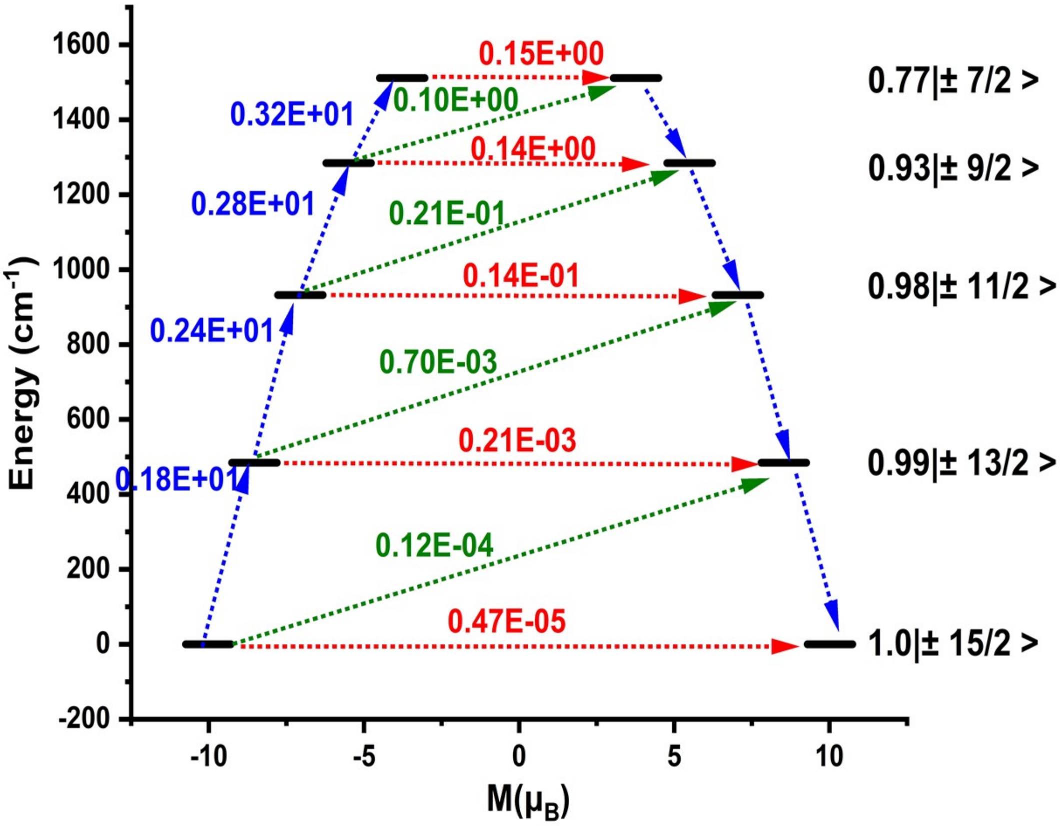

Figure S9: *Ab initio* computed magnetisation blockade barriers for **$1_{Al(OCF3)4}$@F2.**

Table S3. CASSCF+RASSI-SO computed energies of all low-lying KDs, and SINGLE_ANISO computed g-tensors of **1_{cl}@F1**

KDs	**E(cm^{-1})**	g_x	g_y	g_z	ϑ
1_{cl}@F1					
1	0.0	0.000	0.000	19.921	~
2	489.6	0.000	0.000	16.942	0.3
3	935.7	0.042	0.048	14.020	3.3
4	1283.2	0.299	0.315	11.313	15.2
5	1507.7	0.078	0.088	9.083	34.8
6	1640.9	1.102	1.1832	8.924	8.4
7	1750.8	1.310	2.056	3.633	96.4
8	1816.6	0.824	4.082	15.941	81.2
1_{cl}@F2					
1	0.0	0.000	0.000	20.010	~

2	412.9	0.003	0.003	17.061	0.3
3	769.2	0.062	0.071	14.144	4.3
4	1048.6	0.508	0.530	11.348	12.5
5	1250.7	1.312	1.941	8.748	23.4
6	1383.9	3.965	4.534	8.488	69.4
7	1476.3	1.653	3.433	14.395	89.2
8	1579.5	0.242	0.691	18.973	100.3
1$_{\mathrm{Al(OCF3)4}}$**@F2**					
1	0	0.000	0.000	20.027	~
2	484.4	0.000	0.000	17.064	0.5
3	932.16	0.039	0.044	14.125	3.7
4	1284.0	0.405	0.421	11.356	14.6
5	1510.9	0.047	0.635	9.000	33.3
6	1638.3	8.408	6.534	3.951	4.9
7	1732.5	1.212	2.598	15.427	87.9
8	1784.1	0.187	0.321	19.670	92.6

Table S4. Atoms In Molecules (AIM) analysis for **1**$_{\text{x-ray(major)}}$

Critical Points index	**Interaction**	**ρ(r) (a.u.)**	**∇²ρ(r) (a.u.)**	**H(r) (a.u.)**	**Nature**
241	Dy–C (strong)	0.2417	−0.57497	−0.20561	Strong Dy–C bond
266	Dy–C (moderate)	0.03436	+0.01075	+0.00060	Closed-shell Dy–C
206	Dy–C (moderate)	0.02191	+0.00914	+0.00064	Closed-shell Dy–C
233	Dy–C (weak–moderate)	0.01118	+0.03324	−0.00078	Ionic Dy–C
205	Dy···Si (moderate)	0.03448	+0.01167	+0.00066	Electrostatic Dy–Si
249	Dy···Si (weak)	0.01313	+0.04684	+0.00123	Electrostatic Dy–Si
180	Dy···H	0.00224	+0.00644	+0.00035	Anagostic / borderline agostic

Binding Energy Analysis

Encapsulation of $\mathbf{1}_{\mathbf{Cl}}$ was evaluated in both **F1** and **F2**, whereas the bulky counteranion system $\mathbf{1}_{\mathbf{Al(OCF_3)_4}}$ was investigated only in **F2**. The aluminate counteranion was not considered in the **F1**, as structural optimisation of $\mathbf{1}_{\mathbf{Cl}}$**@F1** did not indicate significant equatorial perturbation or reduction of axiality. Counteranion effects were therefore examined specifically in the Cd-MOF system, where chloride-induced distortion was observed. Binding energies were computed using:

$$\Delta E = \{E_{1_{Cl/Al(OCF3)4}\ @F1/F2}\} - \{E_{F1/F2} + E_{1_{Cl/Al(OCF3)4}}\}$$

where, ΔE is the binding energy, $E_{1_{Cl}@F1/F2}$ is the energy of the complex in ZIF/MOF, $E_{F1/F2}$ is the energy of ZIF/MOF, and $E_{1_{Cl/Al(OCF3)4}}$ is the energy of the optimised geometry of the title complex.

$$\% \text{ Dispersion} = (\Delta E_{D3} - \Delta E_{no\ D3})/\Delta E_{D3} \times 100$$

Where:

- ΔE_{D3} = binding energy with dispersion
- $\Delta E_{no\ D3}$ = binding energy without dispersion

Table S5: Binding energy of 1 inside F1 and F2.

System	**Binding energy (ΔE_{D3}) (kJ.mol^{-1})**	**Binding energy($\Delta E_{no\ D3}$) (kJ.mol^{-1})**	**Dispersion Contribution (%)**
$\mathbf{1}_{\mathbf{Cl}}$**@F1**	−357	−233.7	34.5
$\mathbf{1}_{\mathbf{Cl}}$**@F2**	-218.5	-121.9	44
$\mathbf{1}_{\mathbf{Al(OCF3)4}}$**@F2**	-246.2	−74.6	70

Table S6: Lattice Parameters of $\mathbf{1}_{\mathbf{Al(OCF3)4}}$@F2 with and without cell constraints.

Lattice Parameters	$\mathbf{1}_{\mathbf{Al(OCF3)4}}$**@F2 – Fixed Cell**	$\mathbf{1}_{\mathbf{Al(OCF3)4}}$**@F2 – Cell relaxed**
a	11.77745	11.76891
b	25.59363	25.15379
c	25.59368	25.37635
α	120.0001	119.8671
β	90.0000	90.9135
ɣ	90.0000	89.6210

Table S7: Selected structural parameters of $\mathbf{1}_{\mathbf{Al(OCF3)4}}$@F2 with EDIFFG= -0.02 and -0.001 eV.Å^{-1} .

Lattice Parameters	**-0.02**	**-0.001***
Dy-N1	2.229	2.230
Dy-N2	2.196	2.198
N1-Dy-N2	144.5	144.2
Dy-C1	2.751	2.755
Dy-C2	2.696	2.700

C1-C2	1.366	1.366

* Selective dynamics have been performed here for -0.001 eV.Å^{-1}. The MOF is kept fixed.

Origin of Host–Guest Stabilisation in 1$_{Cl}$@F2

To elucidate the nature of host–guest interactions in **1$_{Cl}$@F2**, reduced density gradient (RDG), noncovalent interaction (NCI), and electrostatic potential (ESP) analyses were performed in conjunction with binding energy calculations.

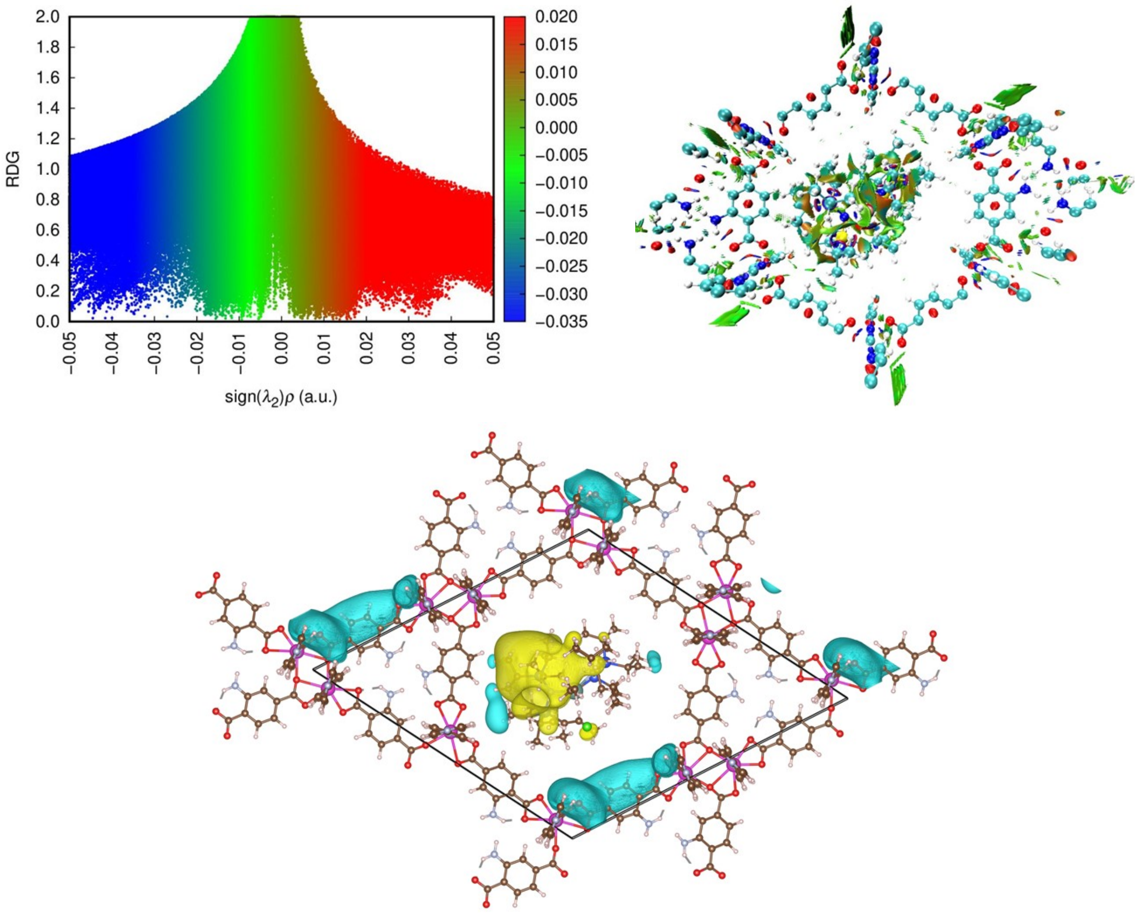

Figure S10: a) RDG scatter plot, b) NCI, and c) ESP difference plot for $\mathbf{1_{Cl}}$**@F2**

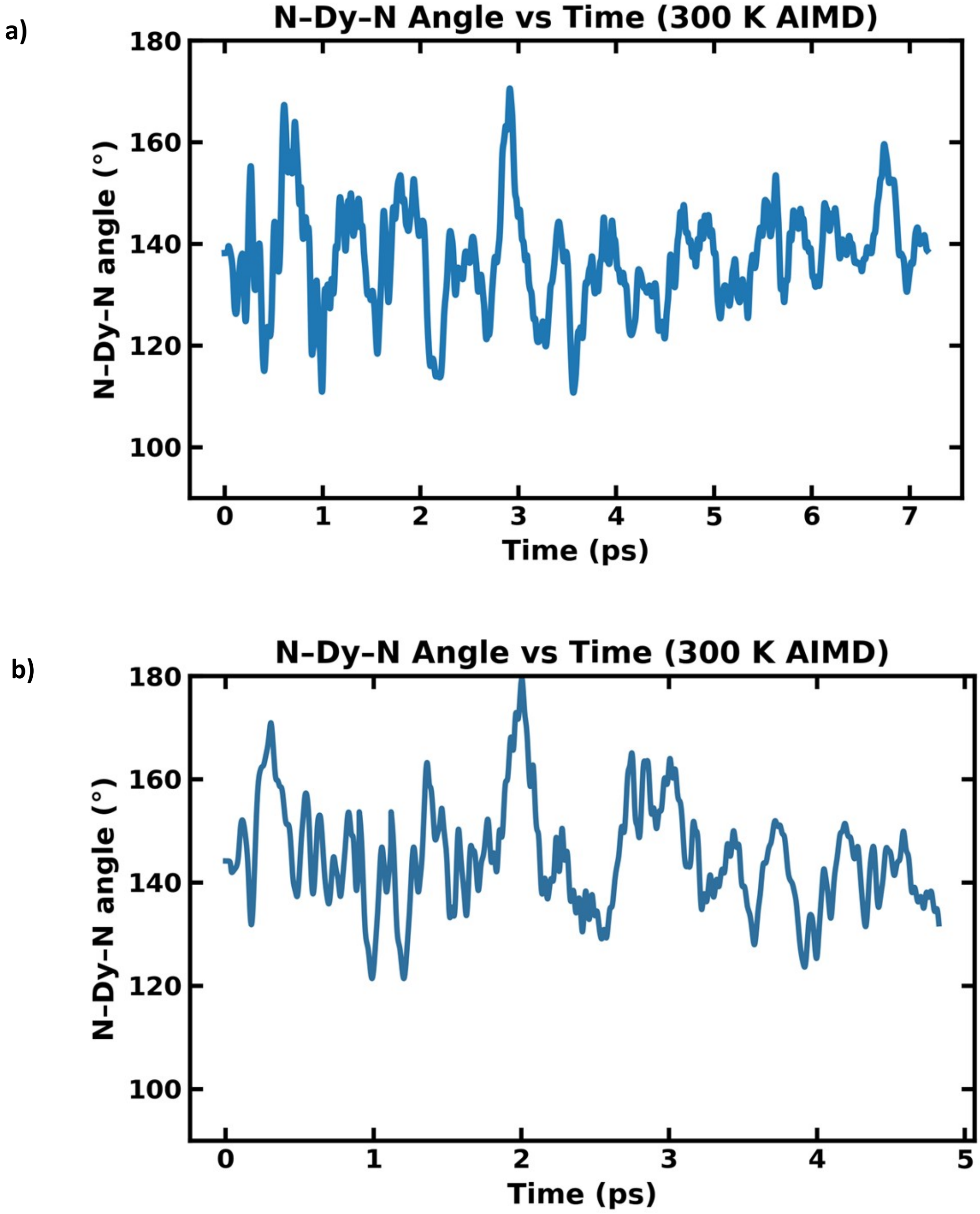

Figure S11: N-Dy–N angle as a function of simulation time at 300 K for a) **1_{Cl}@F2** and b) **$1_{Al(OCF3)4}$@F2.**

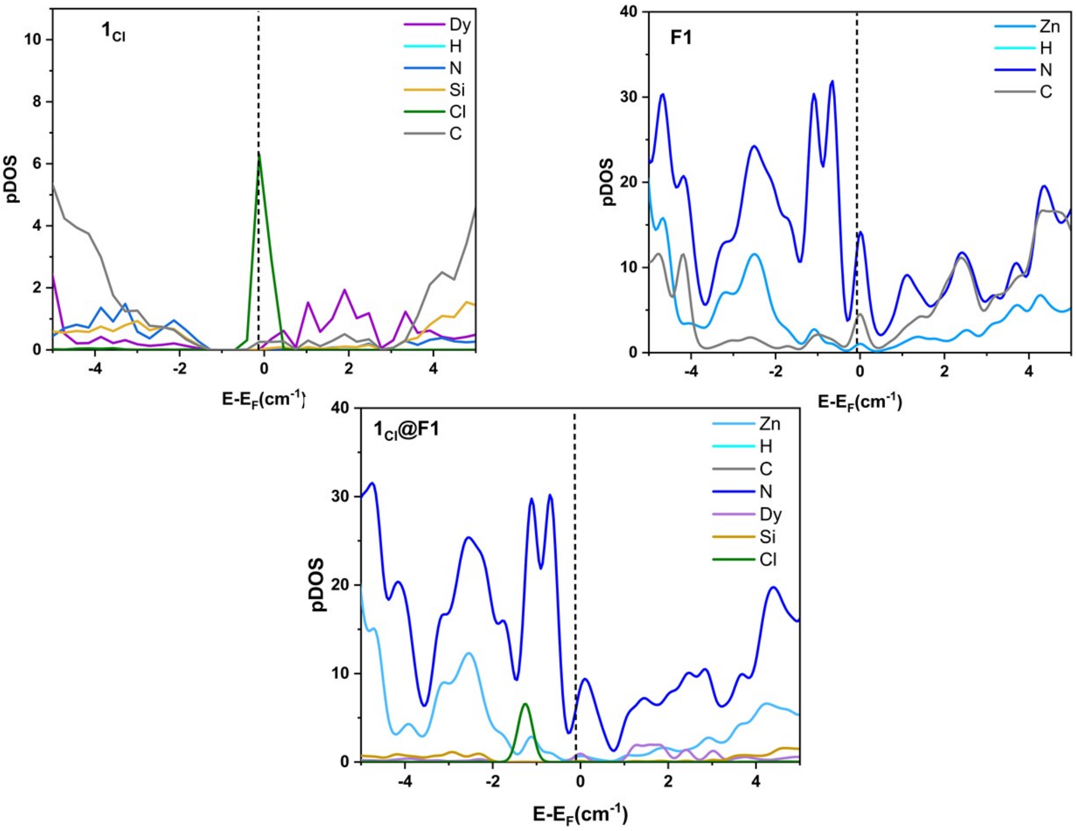

Figure S12: Projected Density of states plot for **1_{Cl}**, **F1** and **1_{Cl}@F1.**

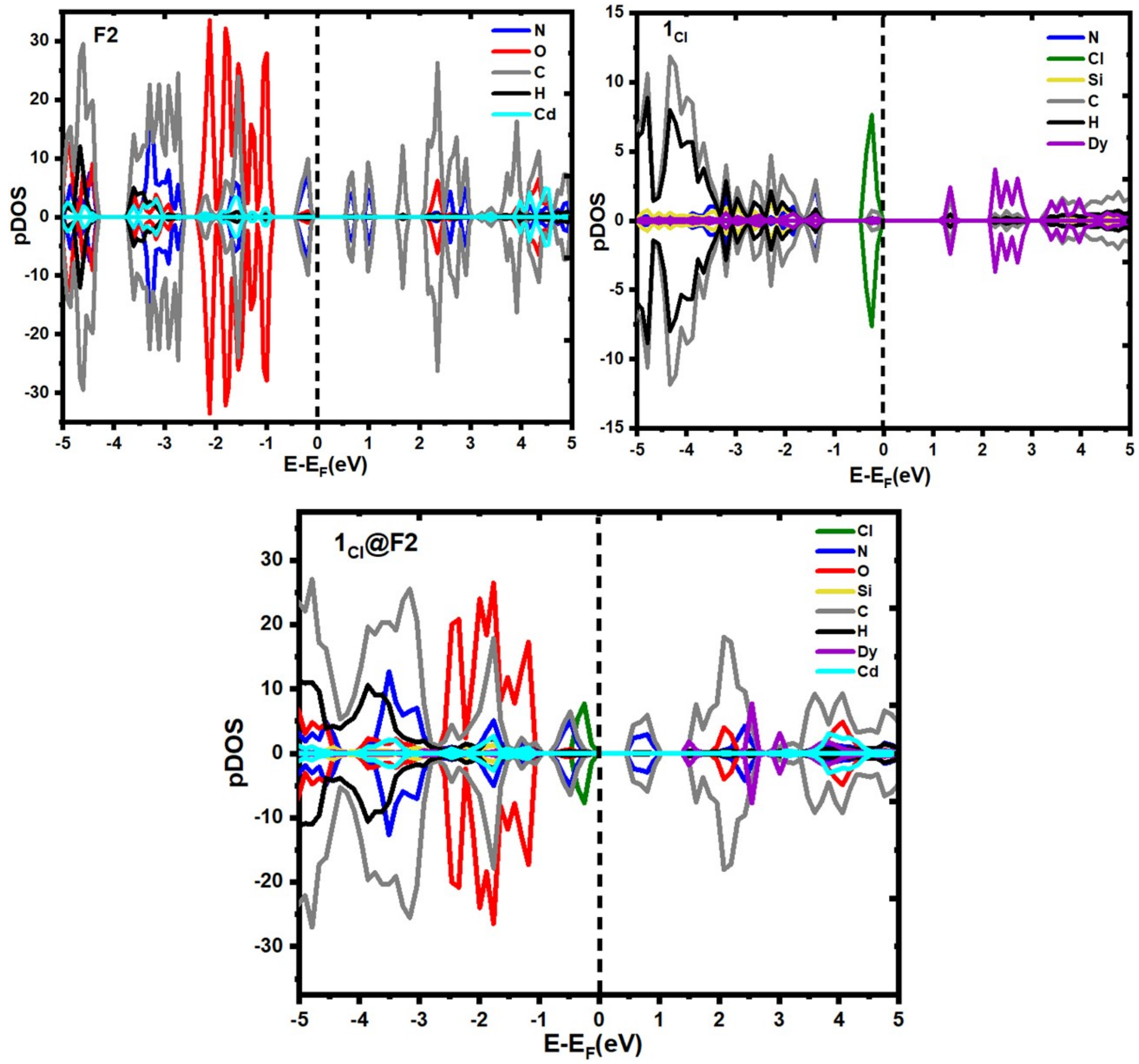

Figure S13: Projected Density of states plot for **F2, 1$_{Cl}$**, and **1$_{Cl}$@F2.**

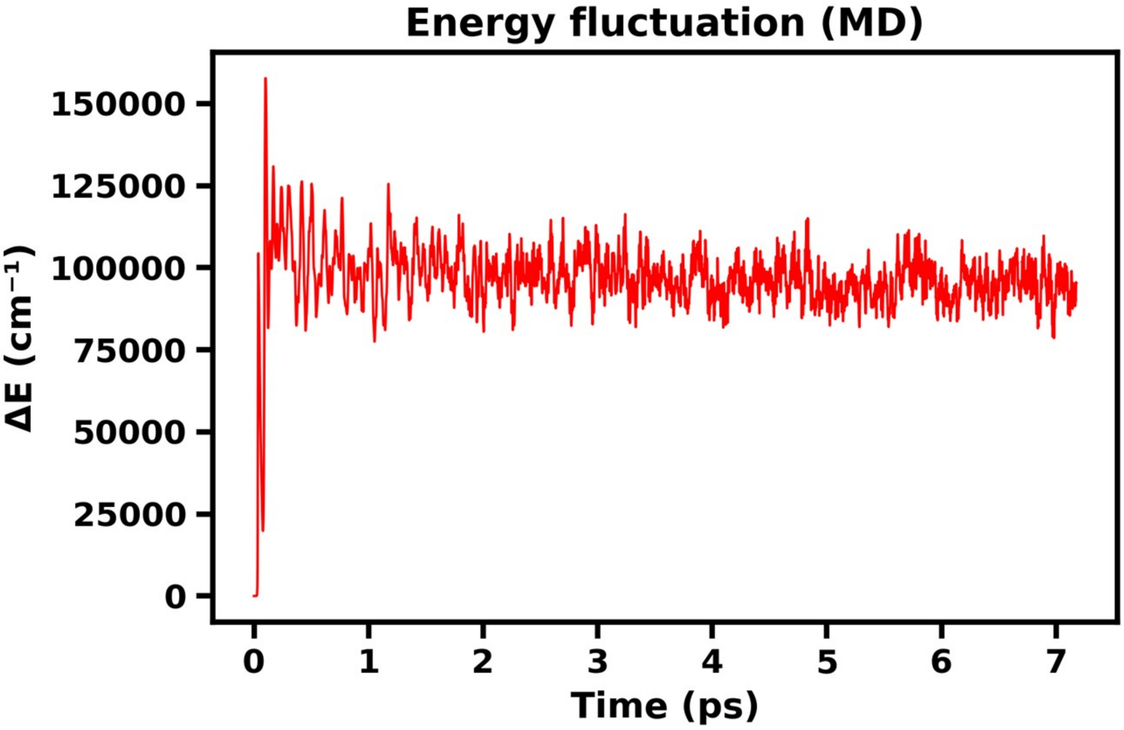

Figure S14: Figure X. Time evolution of the energy (ΔE, in cm⁻¹) during the AIMD simulation at 300 K for $\mathbf{1}_{\mathrm{Cl}}$@**F2**. The system exhibits an initial equilibration phase followed by stable fluctuations around a constant mean value, indicating thermal stability.

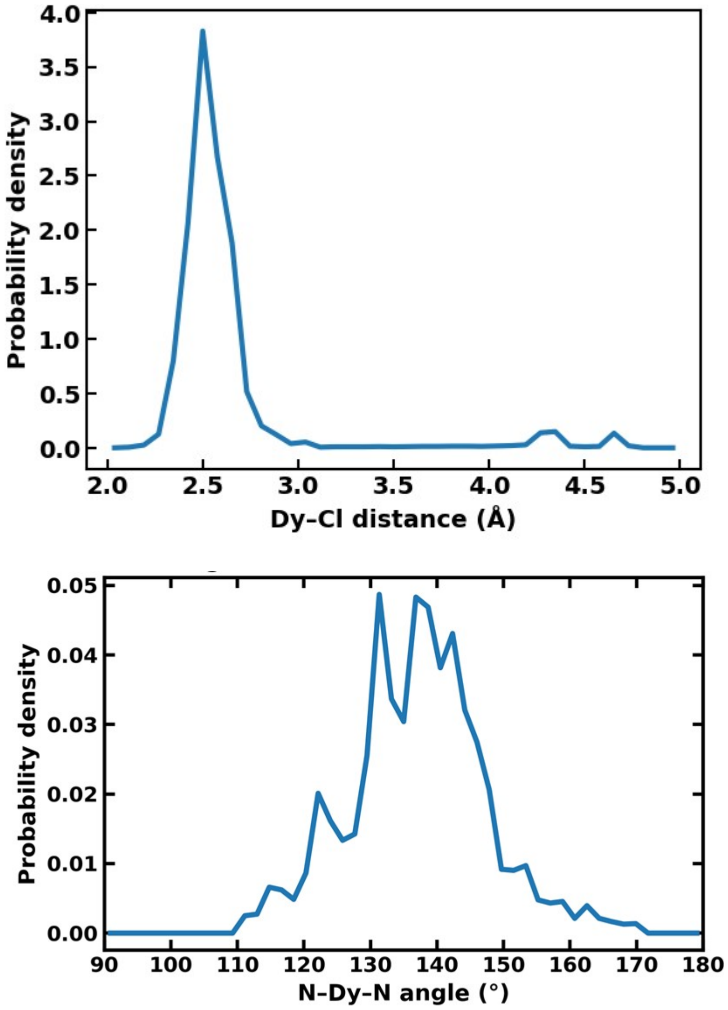

Figure S15: Distributions of (top) Dy–Cl distances and (bottom) N–Dy–N angles from AIMD simulations at 300 K for $\mathbf{1}_{\mathrm{Cl}}$@**F2**.

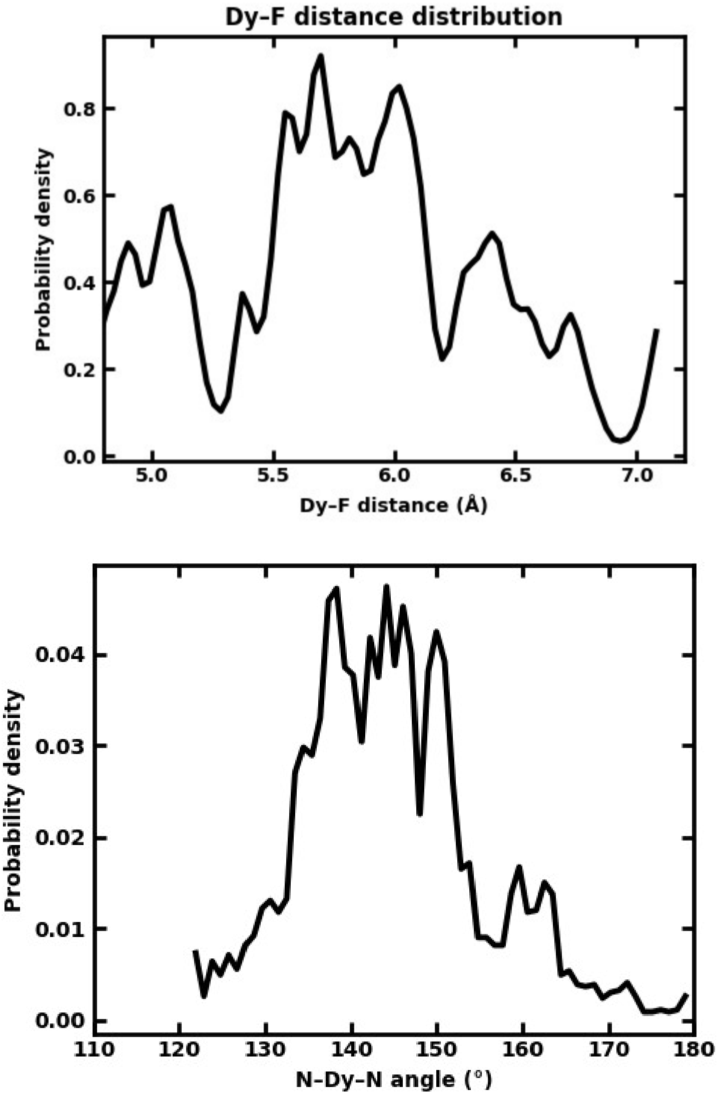

Figure S16: Distributions of (top) Dy–F distances and (bottom) N–Dy–N angles from AIMD simulations at 300 K for **$1_{Al(OCF3)4}$@F2.**

Cartesian Coordinates

$1_{xray\ (major)}$

Dy	4.134380000	14.718008000	6.083398000
N	3.610311000	16.738000000	5.335794000
N	3.517098000	12.726798000	6.801371000
C	6.226761000	12.860573000	5.871425000
C	6.647383000	13.780778000	6.689686000
H	6.115674000	13.976655000	7.450943000
Si	3.910732000	16.689355000	3.644568000
Si	3.367906000	17.986560000	6.516461000

Si	4.828709000	11.660658000	6.536974000
C	3.609600000	17.023790000	8.130191000
H	3.028765000	16.214476000	8.052559000
C	4.158029000	14.780031000	3.375613000
H	5.019055000	14.515746000	3.785656000
H	3.439156000	14.300754000	3.858980000
C	4.154117000	14.309794000	1.914595000
H	4.284658000	13.338654000	1.882890000
H	3.295424000	14.536845000	1.499698000
H	4.879478000	14.752404000	1.426191000
C	5.076024000	16.488693000	8.237317000
H	5.325540000	16.049001000	7.398656000
H	5.685126000	17.235782000	8.410223000
H	5.133561000	15.843334000	8.974071000
C	4.727992000	19.320249000	6.304488000
H	5.488971000	18.828527000	5.880519000
C	6.921908000	12.487626000	4.551722000
H	6.421912000	11.770677000	4.109724000
H	6.953303000	13.273307000	3.965400000
H	7.835070000	12.183411000	4.737209000
C	7.918950000	14.546939000	6.502785000
H	8.027823000	15.183320000	7.240953000
H	8.677243000	13.926287000	6.493189000
H	7.885323000	15.035114000	5.653867000
C	5.530369000	17.528484000	3.138568000
H	5.512693000	18.456168000	3.512302000
C	2.515400000	17.224452000	2.491253000
H	2.770059000	16.930371000	1.569011000
C	1.624099000	18.679754000	6.429848000
H	1.513013000	19.022642000	5.496939000
C	5.569634000	10.732345000	8.002551000

H	4.951628000	9.965231000	8.175890000
C	4.488832000	10.481012000	5.091911000
H	5.398379000	10.468709000	4.677173000
C	3.181480000	17.727119000	9.447615000
H	2.262232000	18.055697000	9.357811000
H	3.230008000	17.089157000	10.188608000
H	3.781572000	18.481606000	9.624783000
C	5.313254000	19.887776000	7.558092000
H	5.534724000	19.157855000	8.173566000
H	6.125181000	20.390484000	7.341081000
H	4.661283000	20.484875000	7.980693000
C	4.345647000	20.376255000	5.271974000
H	3.971254000	19.937759000	4.480221000
H	3.678094000	20.981725000	5.657195000
H	5.142008000	20.887313000	5.017561000
C	6.744525000	16.806914000	3.765370000
H	6.609903000	16.720751000	4.732993000
H	6.835698000	15.915612000	3.368502000
H	7.557681000	17.325857000	3.594310000
C	5.764179000	17.652124000	1.620568000
H	5.001984000	18.108456000	1.207266000
H	6.581790000	18.167905000	1.455708000
H	5.859807000	16.757640000	1.229899000
C	2.347557000	18.752722000	2.443388000
H	3.207969000	19.171030000	2.233193000
H	1.692061000	18.988691000	1.753769000
H	2.032755000	19.073861000	3.315098000
C	1.170718000	16.557607000	2.819469000
H	1.286412000	15.585393000	2.848074000
H	0.854618000	16.874652000	3.692137000
H	0.513908000	16.789483000	2.130807000

C	0.567642000	17.597398000	6.603073000
H	0.779390000	16.839749000	6.017709000
H	0.557271000	17.297298000	7.535071000
H	-0.311609000	17.957940000	6.364729000
C	1.280625000	19.861427000	7.350677000
H	1.954976000	20.563944000	7.247221000
H	0.398691000	20.216354000	7.112265000
H	1.267573000	19.555712000	8.282584000
C	5.670802000	11.484319000	9.315416000
H	4.800760000	11.881588000	9.531629000
H	5.932490000	10.864579000	10.027532000
H	6.342190000	12.193181000	9.235026000
C	6.913963000	10.106039000	7.646984000
H	6.835586000	9.623681000	6.798248000
H	7.589489000	10.810725000	7.562194000
H	7.179806000	9.482124000	8.354700000
C	4.232868000	9.031791000	5.130659000
H	4.725391000	8.628319000	5.871060000
H	3.267685000	8.875437000	5.258617000
H	4.512781000	8.625117000	4.284909000
C	3.696183000	11.074889000	3.927199000
H	3.855941000	12.041428000	3.881819000
H	3.985551000	10.657817000	3.088128000
H	2.740438000	10.908157000	4.061836000
Si	1.975683000	12.404524000	7.510227000
C	-0.332044000	14.291552000	7.350677000
H	-0.211367000	14.455182000	8.310094000
H	-0.662879000	15.107697000	6.920326000
H	-0.979308000	13.568178000	7.221693000
C	1.980173000	12.647750000	9.415705000
H	2.607734000	11.962238000	9.784264000

C	1.016490000	13.892256000	6.726155000
H	1.614907000	14.689652000	6.810260000
C	2.540947000	14.019950000	9.839651000
H	3.393813000	14.179668000	9.385345000
H	1.903018000	14.722954000	9.593694000
H	2.679361000	14.030003000	10.809507000
C	0.637098000	12.459250000	10.165588000
H	0.245703000	11.595230000	9.922571000
H	0.796784000	12.486674000	11.131980000
H	0.020440000	13.179604000	9.916189000
C	0.853296000	13.628761000	5.203596000
H	1.718704000	13.375523000	4.821246000
H	0.209863000	12.901881000	5.066474000
H	0.526293000	14.441399000	4.765108000
C	1.892052000	9.718904000	7.954686000
H	2.868317000	9.776609000	7.900417000
H	1.607147000	9.859367000	8.882102000
H	1.599319000	8.833197000	7.654095000
C	1.265101000	10.795179000	7.061209000
H	1.538078000	10.598064000	6.118248000
C	-0.272391000	10.734372000	7.120470000
H	-0.648986000	11.429857000	6.541373000
H	-0.577049000	9.854624000	6.814431000
H	-0.569205000	10.880794000	8.042439000

1$^{+}_{\text{(opt)}}$

Dy	30.353718818	29.946419398	29.865537104
N	29.717265212	31.904320363	29.112634796
N	29.638011491	28.028626088	30.603733023
C	32.380819436	28.147917937	29.614005675
C	32.878600870	29.066294881	30.494335286
H	32.342602170	29.165777949	31.460785179

Si	30.074891814	31.801312251	27.435951517
Si	29.522878499	33.129297195	30.321234934
Si	30.952850091	26.960387648	30.240468774
C	29.853622319	32.061232137	31.886680134
H	29.237114567	31.129111046	31.725334472
C	30.318698599	29.888173304	27.209421355
H	31.269918707	29.511756049	27.670378844
H	29.474535207	29.312116312	27.673463395
C	30.371380631	29.462532293	25.735763730
H	30.514722206	28.374653960	25.615808292
H	29.434726337	29.732333856	25.219872782
H	31.196536269	29.966995014	25.207443877
C	31.337288617	31.687197930	31.992860044
H	31.854274060	31.493475086	31.019273150
H	31.911788870	32.545927150	32.384389270
H	31.528686792	30.842740013	32.685355338
C	30.840765499	34.491686014	30.183126458
H	31.757586911	33.954257336	29.852273691
C	33.129851418	27.815588689	28.354101253
H	32.461035366	27.538562732	27.521324283
H	33.796991331	28.629329038	28.018706116
H	33.776675517	26.934566937	28.536382745
C	34.158947901	29.834677197	30.416236968
H	34.090425928	30.802858976	30.940679118
H	34.968386370	29.265061761	30.912913947
H	34.483162083	30.019565824	29.378997822
C	31.733860184	32.620629585	27.012066622
H	31.639405309	33.644856407	27.430362858
C	28.705091482	32.326824920	26.250839069
H	29.059356922	32.038366822	25.237813168
C	27.775984136	33.849067154	30.322868854

H	27.669321223	34.253567938	29.291640339
C	31.737425085	26.069420568	31.722593523
H	30.992757305	25.284068940	31.976756707
C	30.701969447	25.715717724	28.828964427
H	31.731519106	25.573175400	28.434634599
C	29.361068423	32.589165324	33.236783707
H	28.266901159	32.722518036	33.244595941
H	29.620995868	31.900097454	34.061927610
H	29.817954251	33.568482063	33.462317125
C	31.166374422	35.220189090	31.492967523
H	31.527944373	34.546323205	32.289448807
H	31.953883531	35.979869390	31.329340868
H	30.286429391	35.752199120	31.895767292
C	30.475431962	35.511340611	29.096306667
H	30.226251438	35.042519050	28.128231005
H	29.598858597	36.115027398	29.394324236
H	31.308020194	36.217684512	28.918540970
C	32.878905701	31.928098791	27.754304376
H	32.669250704	31.830416484	28.840718484
H	33.067030551	30.913464277	27.350624581
H	33.829266472	32.487428051	27.667388601
C	32.037117433	32.751818239	25.516503704
H	31.218641801	33.244515464	24.963398459
H	32.954331902	33.345819080	25.347331222
H	32.206008128	31.767555433	25.043306643
C	28.497383198	33.844499247	26.263800158
H	29.414280697	34.403424803	26.004249321
H	27.715227925	34.139359867	25.539831517
H	28.162473070	34.193447789	27.258419442
C	27.395999946	31.592905578	26.543980789
H	27.499779555	30.494254379	26.482059220

H	27.036662276	31.834125887	27.561727427
H	26.602955813	31.891120038	25.833343251
C	26.721610492	32.755899086	30.499082262
H	26.849619579	31.944056415	29.760229338
H	26.771930667	32.297493616	31.505572617
H	25.698637468	33.161122281	30.384259900
C	27.552108738	35.008471548	31.299499011
H	28.236541922	35.853564338	31.113267240
H	26.520582459	35.397662694	31.210369084
H	27.689295407	34.698672244	32.350658859
C	31.943567156	26.936885478	32.962712156
H	31.044360689	27.521305548	33.226407718
H	32.211107584	26.315028558	33.836954697
H	32.784125123	27.642413254	32.820023201
C	33.042824843	25.374501314	31.318689655
H	32.922777901	24.701132696	30.451361215
H	33.821167134	26.118837659	31.063022783
H	33.440564162	24.767813375	32.152957614
C	30.196564209	24.331773846	29.249244127
H	30.782161118	23.893416200	30.075854701
H	29.142738744	24.364869803	29.571506885
H	30.251710497	23.630695679	28.395981561
C	29.840722238	26.282192322	27.699705825
H	30.228004297	27.242140044	27.314365778
H	29.785939493	25.579193056	26.847785426
H	28.806696367	26.469521766	28.043775568
Si	28.050250358	27.758080831	31.310491441
C	25.747056099	29.562248154	31.250468430
H	25.926997016	29.914614216	32.280316268
H	25.219525168	30.370262778	30.709834387
H	25.055709970	28.703388921	31.311240230

C	28.188358720	27.899379918	33.200143301
H	28.913753236	27.098225402	33.464127538
C	27.044979135	29.180818865	30.537913538
H	27.717534285	30.079071083	30.616849795
C	28.784219836	29.234720013	33.638290559
H	29.755775147	29.433994854	33.144307453
H	28.104179085	30.075083619	33.400103554
H	28.961057207	29.264817368	34.730549796
C	26.891390624	27.615720612	33.967955322
H	26.456748018	26.632621673	33.721697773
H	27.075667055	27.628118660	35.058882272
H	26.116857864	28.374896296	33.762785048
C	26.794927838	28.933170989	29.047175507
H	27.712469816	28.631598878	28.504431754
H	26.063623653	28.118989091	28.897747521
H	26.388817644	29.832733801	28.550061838
C	27.912270612	24.956245198	31.760177746
H	29.016099336	24.964658660	31.792171392
H	27.558254868	25.055942035	32.801771387
H	27.603780907	23.953341206	31.409087207
C	27.328097972	26.058258449	30.866938497
H	27.664423136	25.875458653	29.822698892
C	25.795800247	25.974949600	30.876660087
H	25.321934077	26.664496220	30.157910605
H	25.467011568	24.951834053	30.613251944
H	25.374779695	26.198769982	31.873047436

1$_{Cl}$

H	28.830635053	31.751365363	28.259986045
H	27.713981215	30.690344970	29.245018540
H	32.519930162	29.668353951	33.923275822
H	33.744665191	29.377794434	32.654325655

H	33.665565284	30.957667106	33.476452926
H	28.714453212	29.501569339	32.808530117
H	30.085242105	28.808317779	33.702121152
H	28.560362476	29.136614469	34.539428178
H	32.098455147	31.755135470	30.141704902
H	33.403033136	32.131317187	31.108186755
H	33.430445412	30.524530275	30.346255793
H	28.839955229	31.600468688	34.182048749
H	33.213472290	33.458503267	32.207445035
H	31.815661851	34.454282485	31.685733319
H	32.824669253	34.956837317	33.057286483
H	29.280587348	34.920196375	31.027496629
H	30.871744423	31.981406502	35.664402955
H	29.851660704	30.655057679	36.270930796
H	31.367925342	30.307717503	35.414651543
H	28.125138386	29.022769507	30.038678822
H	26.820491230	28.046672756	30.670665012
H	28.316039759	28.180400810	31.603349831
H	31.516968797	29.743450858	31.677520939
H	28.178541046	29.047098940	27.319150886
H	28.368812883	30.227458248	26.003943186
H	27.612971652	28.666708588	25.674066393
H	30.923025930	28.245621652	31.477373375
H	26.136487894	33.426534837	30.960964470
H	32.558829387	27.324004553	26.055085043
H	29.720036108	33.735534487	34.826381563
H	30.839346919	35.101071547	34.574621219
H	29.753249073	34.683195025	33.305389802
H	33.013634019	30.529159893	28.554013296
H	30.850626300	33.844398377	29.307651095
H	29.734864075	34.313105783	27.984833051

H	30.465463052	35.550362506	29.022901991
H	33.998807043	32.711540536	28.719401850
H	33.616468263	32.887520647	26.991816247
H	32.489645439	33.485260727	28.198257804
H	26.608571985	31.729904010	27.284299037
H	25.895356003	32.404995172	28.771683438
H	27.052079741	33.363452107	27.852815778
H	27.505411307	34.748042914	32.689195420
H	26.055067868	33.963250940	33.388268972
H	27.659242776	33.271972309	33.644989972
H	26.080725456	30.865874112	31.101299773
H	26.836744222	30.964932238	32.703530855
H	25.218422687	31.643263448	32.450120627
H	26.982683125	35.461811385	30.473948161
H	28.171291902	36.479806136	29.716266282
H	27.274583631	35.269062892	28.719414846
H	28.036544683	26.800486298	28.858593538
H	33.117345635	27.820382379	30.219670936
H	33.267481432	27.628374568	31.986448673
H	33.179501106	26.212657281	30.944836749
H	29.721628849	26.357550107	32.535009856
H	31.123711677	25.304654754	32.312376711
H	31.226631771	26.743832420	33.360821912
H	30.024939242	31.549086677	25.652923412
H	30.531011649	32.855795308	26.762470433
H	31.658023525	32.248221285	25.560200596
H	28.537452997	24.662068501	30.196600230
H	28.478179674	25.572027482	31.671068435
H	27.014132047	25.430405239	30.691779836
H	32.079984413	32.858505643	34.300945167
H	30.988150590	28.372243638	23.995503185

H	29.236623129	28.290887075	23.777656168
H	30.013781548	29.844723711	24.090311763
H	33.497394229	27.584098063	28.383098828
H	34.795314109	27.700930259	27.163103218
H	34.179825176	29.157729049	27.937911591
H	29.751582824	27.462224741	26.053154418
H	32.664515114	29.317236256	24.445496286
H	33.660320304	30.187931520	25.610623995
H	34.279271493	28.713612716	24.848508937
H	32.212102831	24.667581863	30.303872021
H	30.549132445	24.109421972	30.373410625
H	31.651132300	23.414198549	29.171025863
H	31.740205332	25.658443888	27.974719076
H	29.292474439	25.424558480	27.074413699
H	30.076756384	23.834345157	27.357065157
H	28.930433426	24.489908660	28.507733550
C	27.981275082	31.752729412	28.999816341
C	33.054708481	30.128933564	33.079328645
C	29.250476193	29.532001263	33.771324467
C	32.785016385	31.254580372	30.863514387
C	29.731223679	30.940104854	34.098331266
C	32.375660205	34.079417794	32.560479739
C	28.891873716	34.489569801	30.058262591
C	30.488696869	30.976820127	35.429232164
C	27.920958929	28.071595002	30.579372400
C	31.424998068	30.922593232	27.224561102
C	32.061839092	30.648328825	32.050097006
C	28.425676648	29.148978122	26.247623763
C	31.249526751	27.211896480	31.223158431
C	26.828547554	32.884337968	31.641800736
C	32.808247542	28.336203992	26.442916520

C	30.385776319	34.220434761	34.089084705
C	32.519902087	31.310082811	27.962512374
C	30.028383852	34.536192727	29.035774885
C	33.181877394	32.647534389	27.980739024
C	26.822010695	32.337861295	28.180669009
C	27.022853188	33.764889207	32.889343605
C	26.213302373	31.524325590	31.981878221
C	27.762496743	35.455937627	29.697359119
C	28.495302046	26.851016775	29.868870109
C	32.774116450	27.229297967	31.084811351
C	30.812722040	26.357850588	32.411831720
C	30.862427623	31.930193746	26.258163843
C	28.107817368	25.573540414	30.635634904
C	31.451323880	33.298358881	33.491793840
C	30.034506474	28.774844509	24.371350754
C	33.868061439	28.176021099	27.533475209
C	29.792862420	28.562432685	25.875302991
C	33.369203563	29.173571730	25.279508817
C	31.365359328	24.358331464	29.672523949
C	30.895899924	25.415990482	28.653542092
C	29.756178494	24.777777000	27.837482803
N	29.718060305	31.878815922	31.264267725
N	30.681783285	28.556046457	28.740612291
Dy	30.195542411	30.572500154	29.600218874
Si	28.398485826	32.792811221	30.614814151
Si	30.368732893	27.024967452	29.546904713
Si	30.679431282	31.778905172	32.703497984
Si	31.148735308	29.013696166	27.125497440
Cl	29.069431839	36.809221719	32.472223505

$\mathbf{1}_{\mathbf{Al(OCF3)4}}$

Dy	13.262188754	14.766779752	14.988239390

N	12.604975499	16.758161904	14.267904167
N	12.527299549	12.856512631	15.762122738
C	15.291186646	13.036422253	14.768291637
C	15.785708450	13.949968857	15.656712013
C	12.692125943	16.936172061	17.071010306
C	13.168692009	14.782286189	12.294198206
C	13.178129099	14.346458075	10.818658161
C	14.177013153	16.564322955	17.220203501
C	13.673412503	19.381053991	15.346167033
C	16.035230840	12.715548312	13.501040754
C	17.066988198	14.718383820	15.601282351
C	14.579836430	17.537297808	12.133280517
C	11.541250129	17.236000550	11.401234387
C	10.612558157	18.683212976	15.440018309
C	14.675954348	10.936794459	16.902508404
C	13.654350056	10.557512878	13.991538262
C	12.159678187	17.484700526	18.402009071
C	13.992696830	20.120734684	16.655705439
C	13.305050566	20.399426053	14.253673144
C	15.738213407	16.885440910	12.899367343
C	14.897793457	17.629271448	10.633224735
C	11.334416026	18.756978020	11.372088716
C	10.220556828	16.510337792	11.681799395
C	9.549846019	17.591875089	15.612009899
C	10.340209542	19.865962509	16.382104595
C	14.869582560	11.799751937	18.153797035
C	15.998091642	10.259399051	16.505545373
C	13.191323194	9.150497796	14.397616488
C	12.782992594	11.099311843	12.852864006
C	8.566273146	14.276781320	16.427868264
C	11.063839432	12.665756334	18.373760160

C	9.880759296	13.938465259	15.713894755
C	11.609227035	14.017514300	18.840009746
C	9.777174224	12.324981524	19.144049509
C	9.637140695	13.710524416	14.214124766
C	10.850937662	9.707750685	16.908963887
C	10.261165217	10.813115128	16.017012629
C	8.726843505	10.696874248	16.008663478
C	23.459935768	11.816372915	12.597671999
C	20.772312029	12.873079879	16.320883789
C	19.797092561	15.068185492	12.286220418
C	19.297396728	10.479636328	12.915516032
H	15.242562480	14.050517705	16.614091838
H	12.098873402	16.001882825	16.901034093
H	14.130339408	14.414967480	12.721161258
H	12.346996639	14.204612801	12.780176702
H	13.312569668	13.261497432	10.712036092
H	12.234897308	14.616820571	10.327467478
H	13.992237217	14.836958924	10.272250223
H	14.706019307	16.364569365	16.264228751
H	14.741729169	17.412463620	17.630885594
H	14.341972525	15.715390053	17.904232978
H	14.596898022	18.866257055	15.021679823
H	15.364850932	12.441224467	12.675947608
H	16.696695117	13.525381114	13.169234357
H	16.688172488	11.844054727	13.669135429
H	16.972990345	15.707559865	16.070699537
H	17.840070190	14.172583428	16.167841966
H	17.452749639	14.840522505	14.583140083
H	14.468966120	18.567731331	12.513201655
H	11.897413676	16.931571729	10.399841580
H	10.525000724	19.063436045	14.404588003

H	13.946113558	10.144353143	17.152669816
H	14.681604606	10.445917892	13.600996389
H	11.070276681	17.612287523	18.379425538
H	12.399352633	16.810340950	19.238666492
H	12.606649956	18.463087912	18.625392619
H	14.378590497	19.456477520	17.440302258
H	14.759101272	20.892941523	16.482204676
H	13.108515093	20.630626669	17.063700121
H	13.058832573	19.927211768	13.293984018
H	12.434662336	21.003204522	14.548918497
H	14.137955807	21.097566253	14.073988273
H	15.531731636	16.817120263	13.981285788
H	15.953509753	15.873793410	12.525428384
H	16.674669248	17.453098387	12.792770752
H	14.081552997	18.089517191	10.058819185
H	15.803271183	18.231914363	10.464330831
H	15.095081883	16.639238049	10.198182829
H	12.255894297	19.301655888	11.122046756
H	10.574716244	19.030785921	10.623114135
H	10.978293001	19.131529352	12.342910812
H	10.322058035	15.416212018	11.634851032
H	9.839786498	16.763034975	12.681912952
H	9.447587029	16.800248925	10.952292526
H	9.700904170	16.762179925	14.908456551
H	9.562443035	17.170844777	16.629116919
H	8.537673169	17.993982169	15.445608749
H	11.026545323	20.705553620	16.212647937
H	9.316329481	20.245134859	16.234999122
H	10.426656390	19.577810481	17.439246405
H	13.968801975	12.367864934	18.423038693
H	15.138746625	11.170602765	19.016470129

H	15.699782524	12.509563358	18.020836107
H	15.901413356	9.604984575	15.628701484
H	16.774010009	11.004062749	16.275190021
H	16.380858680	9.643889412	17.333856489
H	13.781571115	8.730961551	15.223229256
H	12.137148781	9.138147981	14.699372576
H	13.291515079	8.463493892	13.542851985
H	13.155076274	12.056031190	12.463134909
H	12.754747325	10.392150415	12.008826683
H	11.746253604	11.261958191	13.183000064
H	8.732167294	14.620874802	17.457079379
H	8.026481570	15.075747879	15.895806989
H	7.897752396	13.406118956	16.473437735
H	11.814002867	11.895936735	18.633715263
H	10.522437665	14.844929931	15.805163592
H	12.573760851	14.254788388	18.366793327
H	10.907841144	14.834099504	18.610097919
H	11.770598910	14.030077994	19.930228913
H	9.381325454	11.333714824	18.890481313
H	9.968306069	12.332382251	20.229500818
H	8.979862399	13.055536406	18.952349887
H	10.556483439	13.435030089	13.673421751
H	8.921337356	12.893421465	14.049303020
H	9.221013342	14.609318155	13.735040581
H	11.946582681	9.742286350	16.968906033
H	10.466916266	9.779779248	17.936043801
H	10.576385213	8.709926200	16.531241697
H	10.607453636	10.637343054	14.982694588
H	8.251315846	11.382596348	15.295956141
H	8.425116343	9.674530300	15.729168682
H	8.295043769	10.899052219	16.999132075

Si	12.931599321	16.693819663	12.576368833
Si	12.379991080	17.978345098	15.476599532
Si	13.884405719	11.816477003	15.407112976
Si	10.948707542	12.539136318	16.469303646
Al	20.901870394	12.738437190	13.490137636
O	22.271143749	12.347494609	12.492339643
O	21.334651861	12.603434649	15.186391494
O	20.381623810	14.405419123	13.230533421
O	19.488714947	11.755208298	13.089908768
F	24.318836868	12.547148489	13.397100556
F	24.075075968	11.713326862	11.374102661
F	23.448623222	10.541130577	13.129978706
F	19.457677054	12.390308068	16.423192255
F	20.660814799	14.230979239	16.585101713
F	21.444116316	12.332239903	17.383867729
F	20.086673955	14.635045368	11.010342427
F	20.091547253	16.411673416	12.319375730
F	18.391111550	15.011255303	12.371908291
F	19.195342812	10.119454813	11.590100126
F	18.103325900	10.053433999	13.499599074
F	20.279117329	9.680217506	13.458256818

INCAR:

References-

1. T. D. Kühne, M. Iannuzzi, M. Del Ben, V. V. Rybkin, P. Seewald, F. Stein, T. Laino, R. Z. Khaliullin, O. Schütt and F. Schiffmann, *J. Chem. Phys.*, 2020, **152**, 194103.
2. J. Hafner, *J. Comput. Chem.*, 2008, **29**, 2044-2078.
3. A. E. Mattsson, R. Armiento, P. A. Schultz and T. R. Mattsson, *Phys. Rev. B Condens.*, 2006, **73**, 195123.
4. E. Caldeweyher, C. Bannwarth and S. Grimme, *Chem. Phys*, 2017, **147**.
5. F. Aquilante, J. Autschbach, R. K. Carlson, L. F. Chibotaru, M. G. Delcey, L. De Vico, I. Fdez. Galván, N. Ferré, L. M. Frutos and L. Gagliardi, *Journal*, 2016.
6. J. Emerson-King, G. K. Gransbury, B. E. Atkinson, W. J. Blackmore, G. F. Whitehead, N. F. Chilton and D. P. Mills, *Nature*, 2025, 1-5.
7. B. O. Roos, P. Linse, P. E. Siegbahn and M. R. Blomberg, *Chem. Phys.*, 1982, **66**, 197-207.