Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

The coexistence of long τ_{QTM} and high U_{eff} as a concise criterion for good single-molecule magnet. A theoretical case study of square antiprism dysprosium single-ion magnets

Hong Liu,[a] Jin-Feng Li [b] and Bing Yin [a]*

[a] Key Laboratory of Synthetic and Natural Functional Molecule of the Ministry of Education, Lab of Theoretical Molecular Magnetism, College of Chemistry and Materials Science, Northwest University, Xi'an, 710127 P. R. China

[b] College of Chemistry and Chemical Engineering, Shaanxi Key Laboratory of Chemical Reaction Engineering, Yan'an University, Yan'an, 716000, P. R. China

* Corresponding author, E-mail: rayinyin@nwu.edu.cn

TABLE S1 *ab initio* calculated principal g values and energies of the lowest-lying KDs of WALQAE

WALQAE	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	6.165E-05	2.821E-03	1.496E-01	3.570E+00	4.724E+00	4.884E-01	5.640E-01	1.867E-01
g_Y	8.793E-05	3.066E-03	1.875E-01	5.816E+00	2.866E-01	3.021E+00	2.967E+00	5.994E-01
g_Z	1.990E+01	1.707E+01	1.399E+01	9.321E+00	9.023E+00	1.035E+01	1.425E+01	1.938E+01
E (cm^{-1})	0.000E+00	2.591E+02	4.784E+02	6.123E+02	6.661E+02	7.420E+02	7.602E+02	7.963E+02

TABLE S2 *ab initio* calculated principal g values and energies of the lowest-lying KDs of VORCUD

VORCUD	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	1.002E-04	3.163E-02	5.033E-01	5.937E-01	1.490E+00	1.137E+00	1.705E+00	1.001E-01
g_Y	1.798E-04	3.268E-02	9.897E-01	2.347E+00	3.682E+00	1.975E+00	2.910E+00	1.653E-01
g_Z	1.994E+01	1.706E+01	1.341E+01	1.630E+01	1.136E+01	9.433E+00	1.467E+01	1.958E+01
E (cm^{-1})	0.000E+00	2.887E+02	5.626E+02	6.529E+02	7.238E+02	7.564E+02	8.054E+02	9.666E+02

TABLE S3 *ab initio* calculated principal g values and energies of the lowest-lying KDs of WALPUX

WALPUX	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	1.272E-03	8.366E-02	2.984E-01	1.572E+00	5.236E+00	2.992E+00	8.965E-01	9.615E-02
g_Y	1.406E-03	8.691E-02	3.245E-01	2.296E+00	3.652E-01	5.217E+00	2.047E+00	2.034E-01
g_Z	1.983E+01	1.673E+01	1.323E+01	1.022E+01	6.833E+00	1.019E+01	1.774E+01	1.959E+01
E (cm^{-1})	0.000E+00	2.021E+02	3.470E+02	4.351E+02	4.911E+02	5.331E+02	5.657E+02	6.091E+02

TABLE S4 *ab initio* calculated principal g values and energies of the lowest-lying KDs of ISEBIS

ISEBIS	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	3.700E-03	1.135E-01	1.028E+00	3.741E+00	3.440E+00	7.341E-01	1.345E-02	9.160E-03
g_Y	5.542E-03	1.913E-01	1.410E+00	5.168E+00	3.622E+00	1.141E+00	1.060E-01	2.154E-02
g_Z	1.946E+01	1.579E+01	1.250E+01	8.625E+00	1.105E+01	1.787E+01	1.835E+01	1.937E+01
E (cm^{-1})	0.000E+00	1.300E+02	1.961E+02	2.443E+02	2.861E+02	3.356E+02	4.186E+02	5.063E+02

TABLE S5 *ab initio* calculated principal g values and energies of the lowest-lying KDs of EYOKIO

EYOKIO	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	6.534E-04	7.727E-02	1.071E-01	1.399E+00	8.965E-01	3.625E+00	1.472E+00	4.338E-01
g_Y	9.508E-04	7.897E-02	1.277E-01	1.891E+00	3.476E+00	3.827E+00	6.182E+00	7.095E-01
g_Z	1.985E+01	1.673E+01	1.317E+01	1.049E+01	7.996E+00	5.921E+00	1.139E+01	1.768E+01
E (cm^{-1})	0.000E+00	2.427E+02	4.236E+02	5.267E+02	5.891E+02	6.263E+02	6.525E+02	6.804E+02

TABLE S6 *ab initio* calculated principal g values and energies of the lowest-lying KDs of DARTUN

DARTUN	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	5.484E-03	2.178E-01	1.968E+00	1.866E+00	2.671E+00	1.073E+00	1.647E-01	1.907E-02
g_Y	1.098E-02	3.582E-01	3.086E+00	5.516E+00	3.993E+00	1.907E+00	4.783E-01	3.845E-02
g_Z	1.948E+01	1.542E+01	1.141E+01	1.135E+01	1.073E+01	1.700E+01	1.908E+01	1.973E+01
E (cm^{-1})	0.000E+00	1.450E+02	2.147E+02	2.476E+02	2.892E+02	3.418E+02	4.008E+02	4.966E+02

TABLE S7 *ab initio* calculated principal g values and energies of the lowest-lying KDs of DARTOH

DARTOH	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	6.155E-03	4.323E-01	2.830E+00	5.550E+00	4.585E-01	1.159E+00	1.281E-02	2.186E-03
g_Y	1.883E-02	5.808E-01	4.786E+00	1.802E+00	3.343E+00	3.610E+00	3.066E-02	7.522E-03
g_Z	1.934E+01	1.581E+01	1.132E+01	7.586E+00	9.449E+00	1.559E+01	1.949E+01	1.986E+01
E (cm^{-1})	0.000E+00	1.196E+02	1.815E+02	2.174E+02	2.588E+02	2.854E+02	3.981E+02	5.389E+02

TABLE S8 *ab initio* calculated principal g values and energies of the lowest-lying KDs of UCIBAL

UCIBAL	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	4.722E-03	7.536E-01	2.958E+00	5.391E+00	6.902E-01	8.837E-01	8.016E-02	1.202E-02
g_Y	1.014E-02	8.436E-01	3.775E+00	3.420E+00	2.713E+00	1.439E+00	1.841E-01	2.436E-02
g_Z	1.962E+01	1.573E+01	1.148E+01	7.360E+00	1.258E+01	1.588E+01	1.881E+01	1.967E+01
E (cm^{-1})	0.000E+00	1.248E+02	1.673E+02	2.034E+02	2.324E+02	2.773E+02	3.809E+02	5.097E+02

TABLE S9 *ab initio* calculated principal g values and energies of the lowest-lying KDs of EYOKEK

EYOKEK	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	6.365E-04	1.844E-01	1.376E+00	6.154E+00	2.846E+00	1.113E-01	2.816E-01	1.549E-02
g_Y	1.734E-03	2.072E-01	1.529E+00	3.623E+00	3.754E+00	8.491E-01	8.898E-01	4.413E-02
g_Z	1.982E+01	1.661E+01	1.287E+01	8.300E+00	1.257E+01	1.761E+01	1.914E+01	1.987E+01
E (cm^{-1})	0.000E+00	1.961E+02	3.365E+02	4.232E+02	4.774E+02	5.425E+02	5.630E+02	6.213E+02

TABLE S10 *ab initio* calculated principal g values and energies of the lowest-lying KDs of GAQGOX

GAQGOX	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	3.080E-03	1.344E-01	1.257E+00	3.421E+00	4.894E-01	2.473E+00	5.334E-01	7.866E-03
g_Y	4.899E-03	1.669E-01	1.360E+00	4.964E+00	5.183E+00	3.635E+00	1.662E+00	1.321E-02
g_Z	1.960E+01	1.623E+01	1.252E+01	8.707E+00	1.081E+01	1.245E+01	1.757E+01	1.986E+01
E (cm^{-1})	0.000E+00	1.438E+02	2.272E+02	2.792E+02	3.128E+02	3.471E+02	3.862E+02	5.282E+02

TABLE S11 *ab initio* calculated principal g values and energies of the lowest-lying KDs of WALQEI

WALQEI	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	7.525E-05	1.607E-01	1.247E+00	2.363E+00	3.938E+00	1.026E+00	5.760E-02	1.419E-02
g_Y	3.353E-04	1.708E-01	1.285E+00	4.661E+00	4.533E+00	1.346E+00	3.355E-01	4.005E-01
g_Z	1.980E+01	1.652E+01	1.288E+01	8.927E+00	1.109E+01	1.780E+01	1.934E+01	1.950E+01
E (cm^{-1})	0.000E+00	1.941E+02	3.299E+02	4.173E+02	4.721E+02	5.330E+02	5.733E+02	5.925E+02

TABLE S12 *ab initio* calculated principal g values and energies of the lowest-lying KDs of EYOKAG

EYOKAG	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	6.608E-04	1.876E-01	1.194E+00	2.670E+00	3.398E+00	3.447E-02	3.229E-01	1.890E-02
g_Y	8.268E-04	2.097E-01	1.278E+00	5.080E+00	4.903E+00	8.869E-01	1.051E+00	7.588E-02
g_Z	1.975E+01	1.645E+01	1.277E+01	8.764E+00	1.211E+01	1.760E+01	1.907E+01	1.980E+01
E (cm^{-1})	0.000E+00	1.732E+02	2.959E+02	3.747E+02	4.230E+02	4.941E+02	5.141E+02	5.595E+02

TABLE S13 *ab initio* calculated principal g values and energies of the lowest-lying KDs of GUYRAU

GUYRAU	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	5.058E-03	2.165E-01	1.844E+00	4.428E+00	2.481E+00	1.916E-02	5.321E-02	1.040E-02
g_Y	6.462E-03	3.563E-01	2.540E+00	6.437E-01	6.405E+00	1.729E-01	8.878E-02	2.922E-02
g_Z	1.957E+01	1.583E+01	1.154E+01	7.234E+00	1.245E+01	1.587E+01	1.851E+01	1.900E+01
E (cm^{-1})	0.000E+00	1.579E+02	2.399E+02	2.955E+02	3.238E+02	4.202E+02	4.791E+02	5.323E+02

TABLE S14 *ab initio* calculated principal g values and energies of the lowest-lying KDs of GAQGOX01

GAQGOX01	KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X	1.787E-02	2.803E-01	7.901E-01	2.721E+00	3.064E-02	2.168E+00	2.720E-01	4.235E-03
g_Y	3.209E-02	4.095E-01	8.497E-01	5.464E+00	3.569E+00	4.219E+00	6.967E-01	7.089E-03
g_Z	1.944E+01	1.592E+01	1.258E+01	8.402E+00	1.058E+01	1.396E+01	1.918E+01	1.990E+01
E (cm^{-1})	0.000E+00	1.212E+02	2.059E+02	2.554E+02	2.943E+02	3.341E+02	4.018E+02	5.135E+02

TABLE S15 *ab initio* calculated principal g values and energies of the lowest-lying KDs of UCEZUZ

UCEZUZ KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X 2.960E-03	6.543E-02	2.014E+00	4.369E+00	3.162E-01	7.204E-01	1.890E-02	3.353E-02
g_Y 6.809E-03	1.083E+00	3.054E+00	6.393E+00	1.765E+00	1.232E+00	2.140E-01	1.255E-01
g_Z 1.968E+01	1.613E+01	1.245E+01	9.204E+00	1.357E+01	1.599E+01	1.816E+01	1.904E+01
E (cm^{-1}) 0.000E+00	1.368E+02	1.530E+02	1.937E+02	2.279E+02	2.854E+02	3.818E+02	4.430E+02

TABLE S16 *ab initio* calculated principal g values and energies of the lowest-lying KDs of DOGMUJ

DOGMUJ KD0	KD1	KD2	KD3	KD4	KD5	KD6	KD7
g_X 2.165E-02	1.595E+00	1.131E+00	6.433E+00	1.989E+00	7.536E-01	6.229E-02	1.742E-02
g_Y 3.247E-02	4.570E+00	3.433E+00	3.026E+00	3.053E+00	1.413E+00	1.288E-01	3.806E-02
g_Z 1.935E+01	1.350E+01	9.947E+00	7.919E+00	1.160E+01	1.652E+01	1.947E+01	1.973E+01
E (cm^{-1}) 0.000E+00	9.648E+01	1.214E+02	1.680E+02	1.991E+02	2.446E+02	3.199E+02	4.268E+02

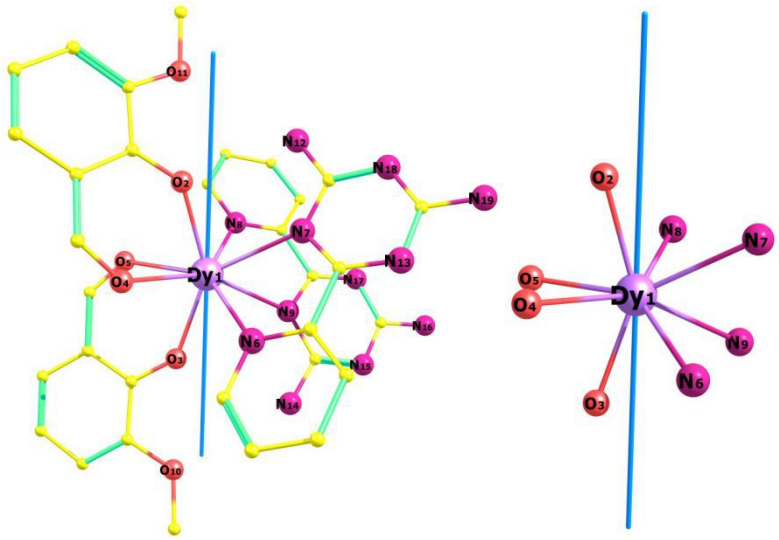

FIG. S1 Structure and *ab initio* magnetic easy axis of WALPUX (left: full, right: first sphere)

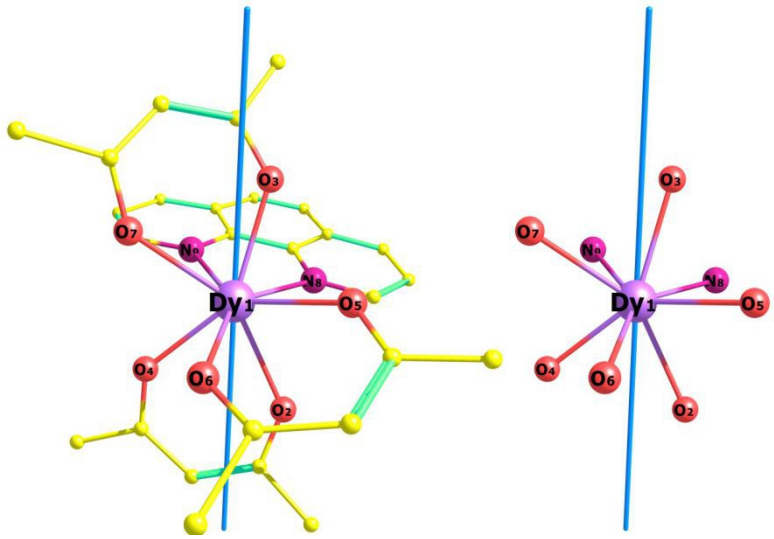

FIG. S2 Structure and *ab initio* magnetic easy axis of ISEBIS (left: full, right: first sphere)

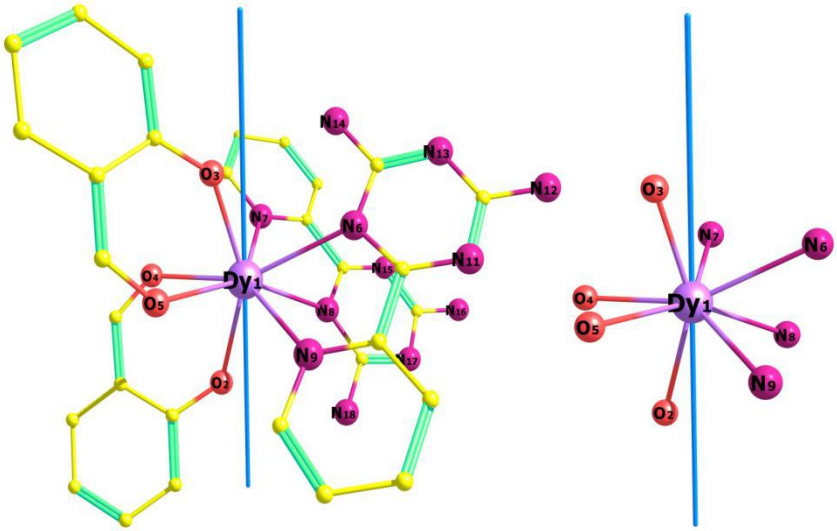

FIG. S3 Structure and *ab initio* magnetic easy axis of EYOKIO (left: full, right: first sphere)

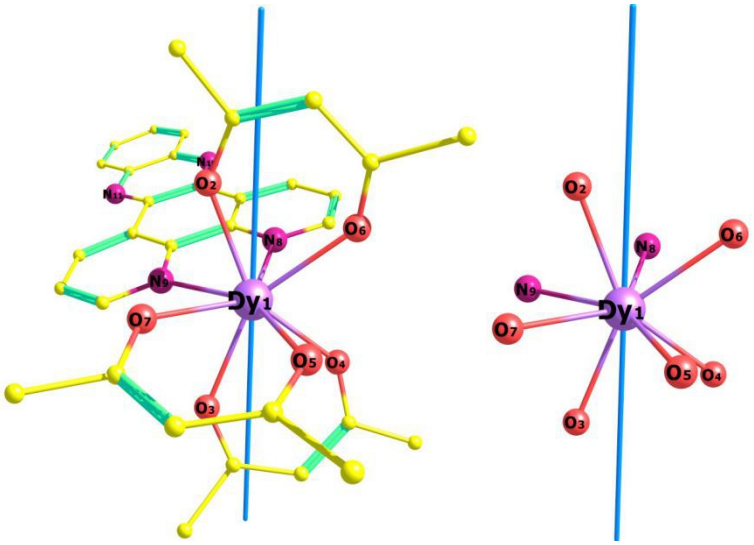

FIG. S4 Structure and *ab initio* magnetic easy axis of DARTUN (left: full, right: first sphere)

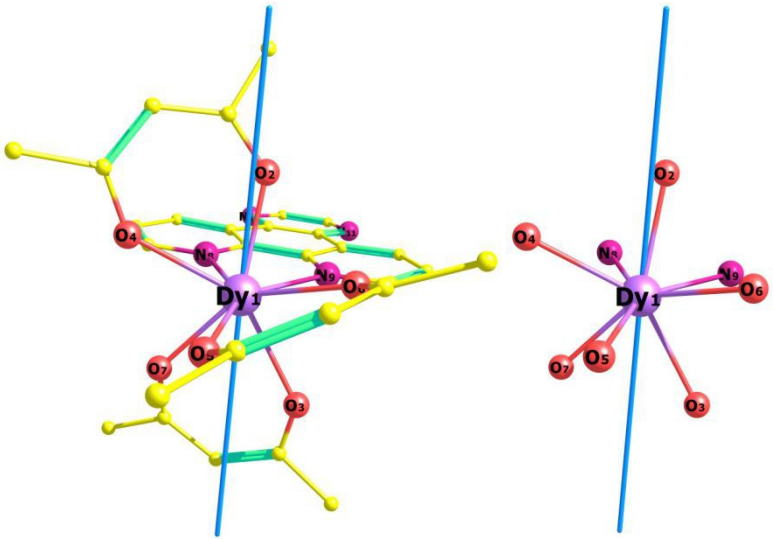

FIG. S5 Structure and *ab initio* magnetic easy axis of DARTOH (left: full, right: first sphere)

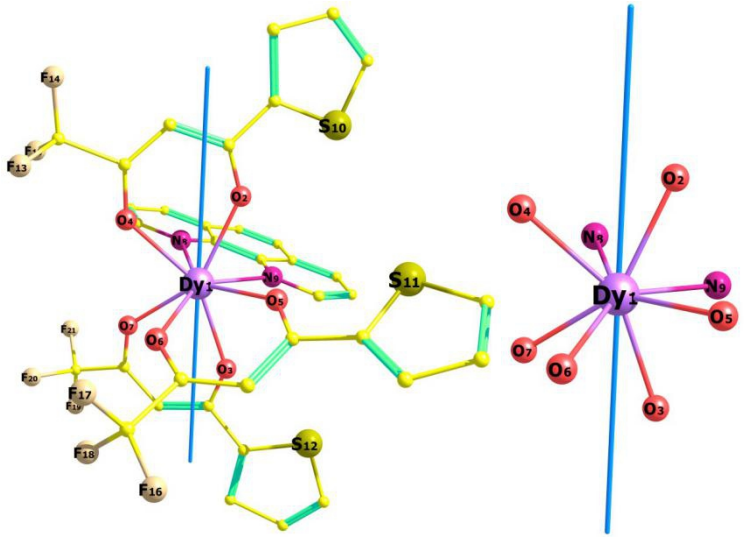

FIG. S6 Structure and *ab initio* magnetic easy axis of UCIBAL (left: full, right: first sphere)

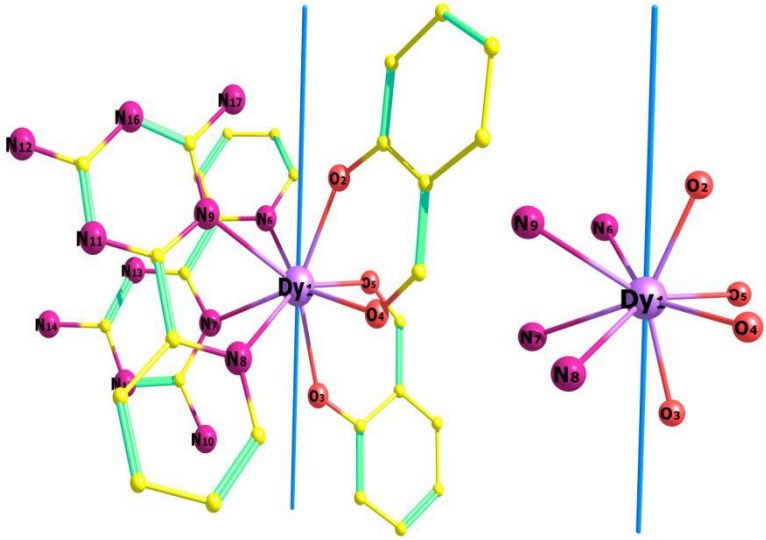

FIG. S7 Structure and *ab initio* magnetic easy axis of EYOKEK (left: full, right: first sphere)

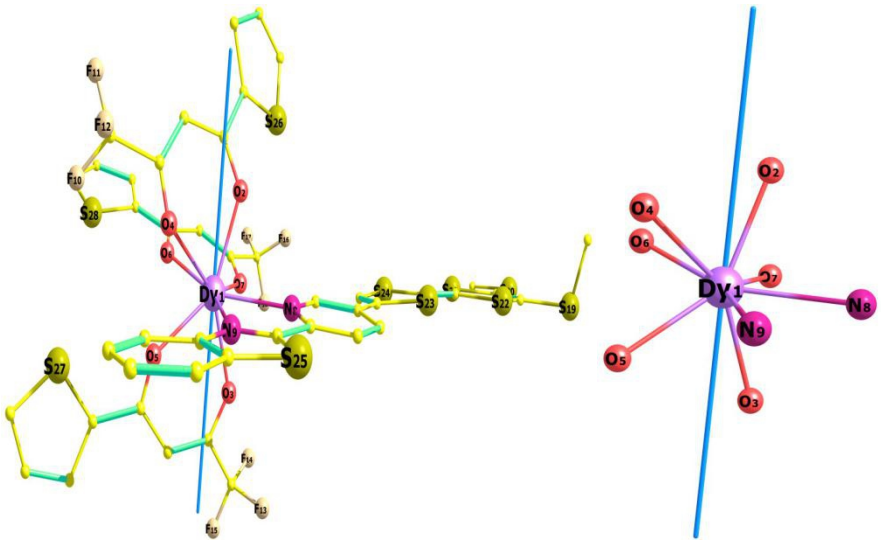

FIG. S8 Structure and *ab initio* magnetic easy axis of GAQGOX (left: full, right: first sphere)

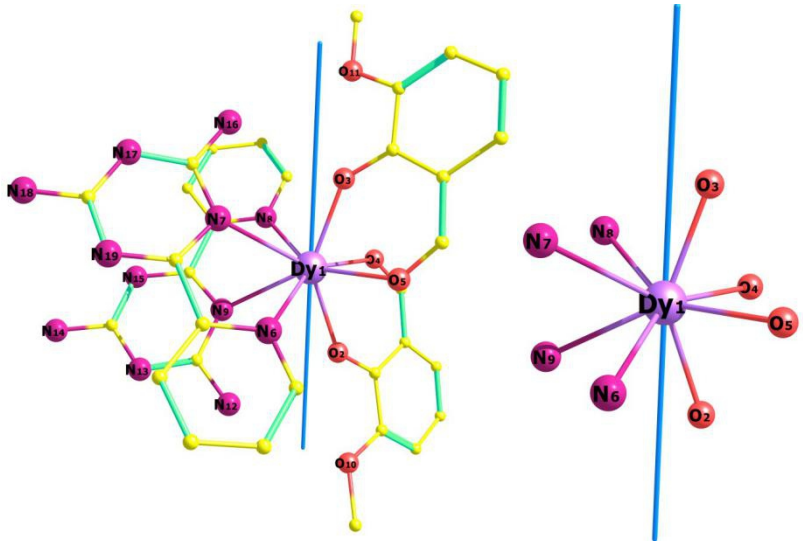

FIG. S9 Structure and *ab initio* magnetic easy axis of WALQEI (left: full, right: first sphere)

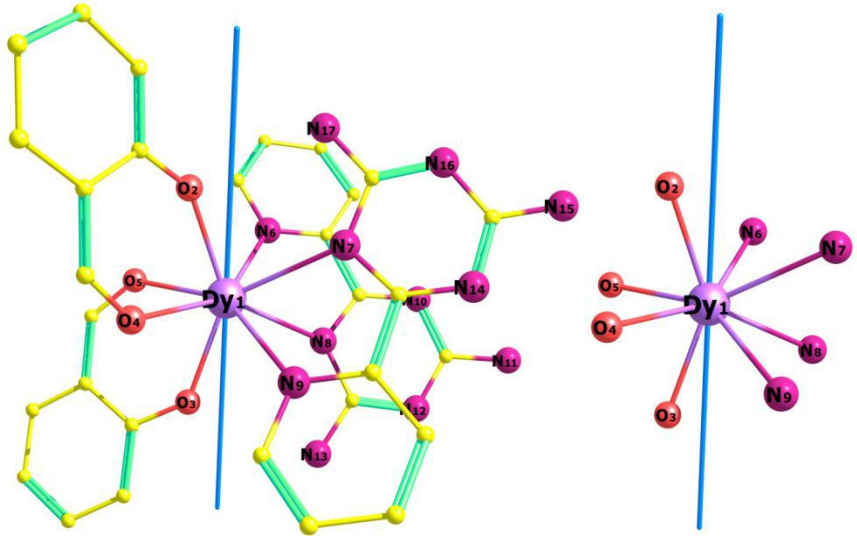

FIG. S10 Structure and *ab initio* magnetic easy axis of EYOKAG (left: full, right: first sphere)

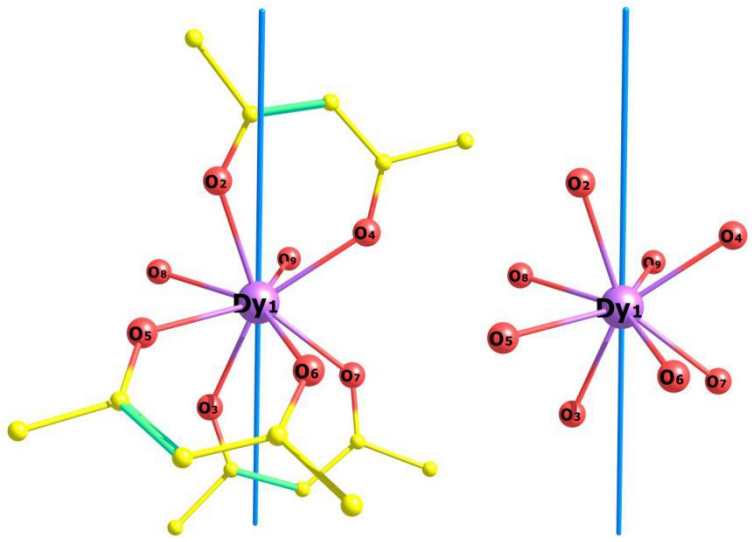

FIG. S11 Structure and *ab initio* magnetic easy axis of GUYRAU (left: full, right: first sphere)

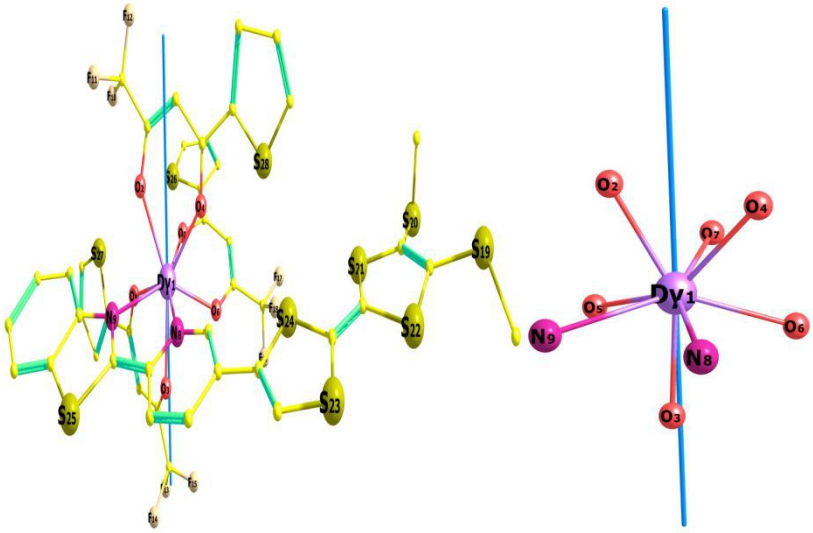

FIG. S12 Structure and *ab initio* magnetic easy axis of GAQGOX01 (left:full, right: first sphere)

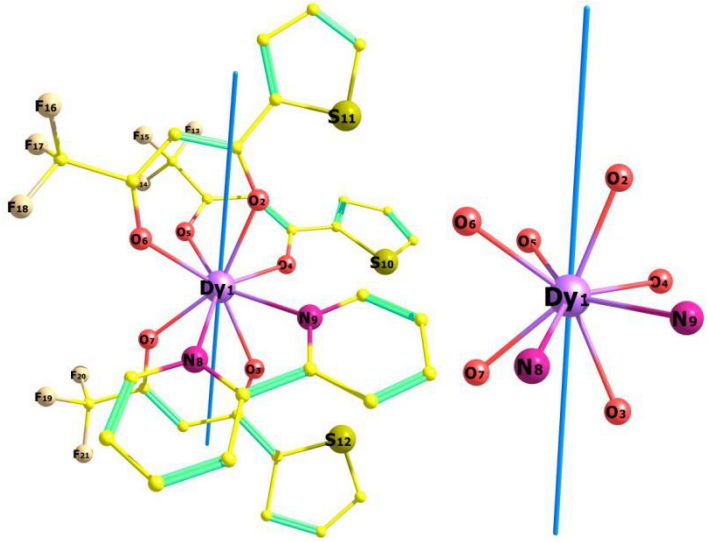

FIG. S13 Structure and *ab initio* magnetic easy axis of UCEZUZ (left: full, right: first sphere)

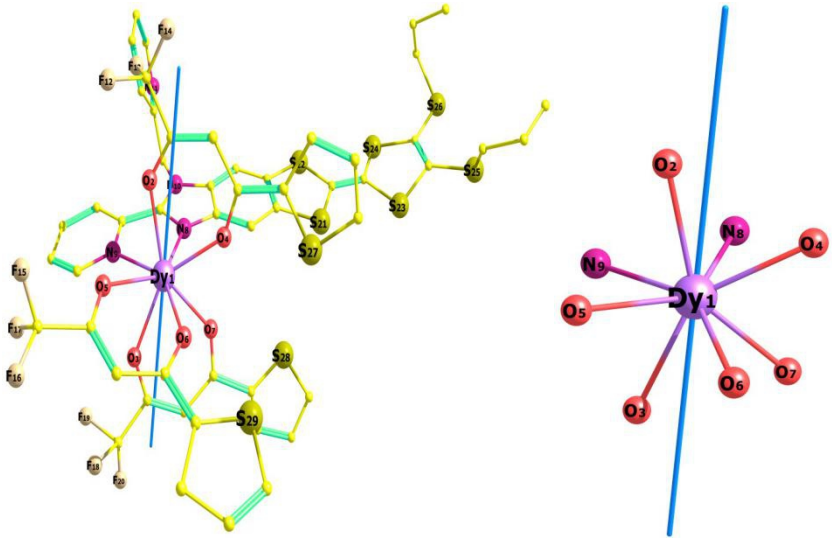

FIG. S14 Structure and *ab initio* magnetic easy axis of DUGMUJ (left: full, right: first sphere)

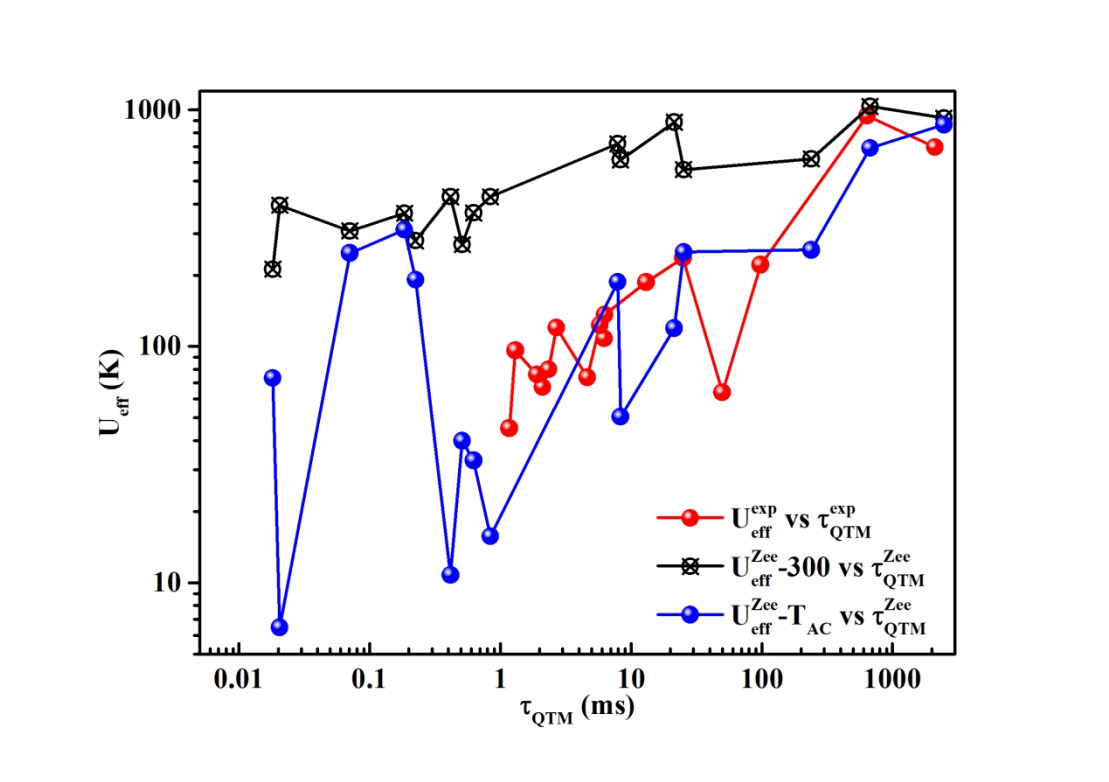

FIG. S15 U_{eff} vs τ_{QTM} for all of the selected SAP Dy-SIM

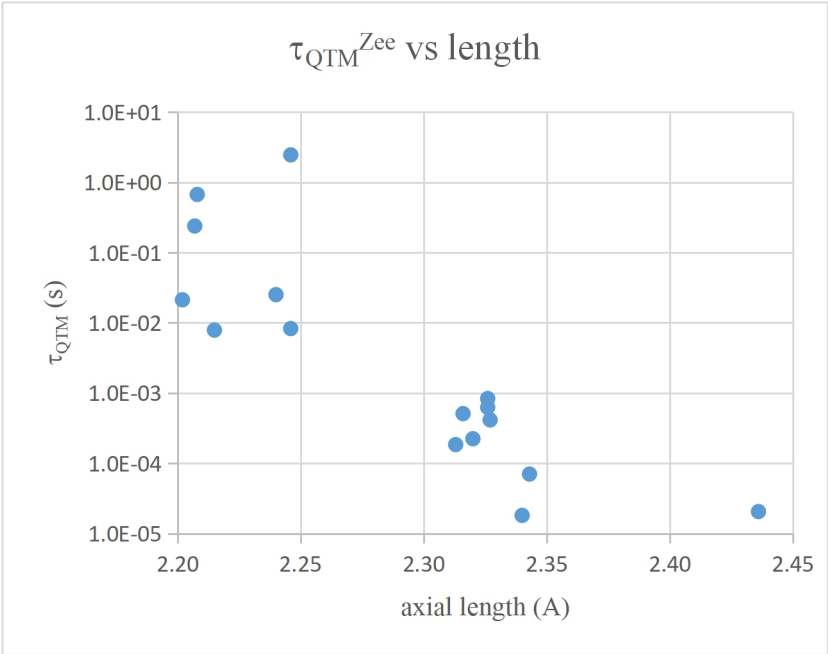

FIG. S16 The relation between τ_{QTM}^{Zee} and averaged bond length of the two axial ligating atoms

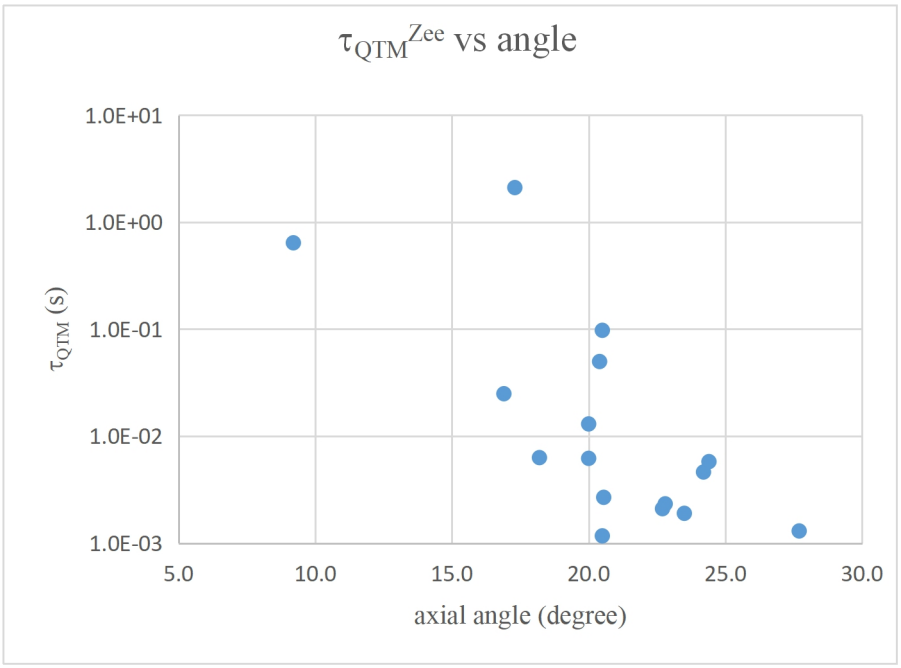

FIG. S17 The relation between τ_{QTM}^{Zee} and averaged angle of the two axial ligating atoms with respect to magnetic easy axis

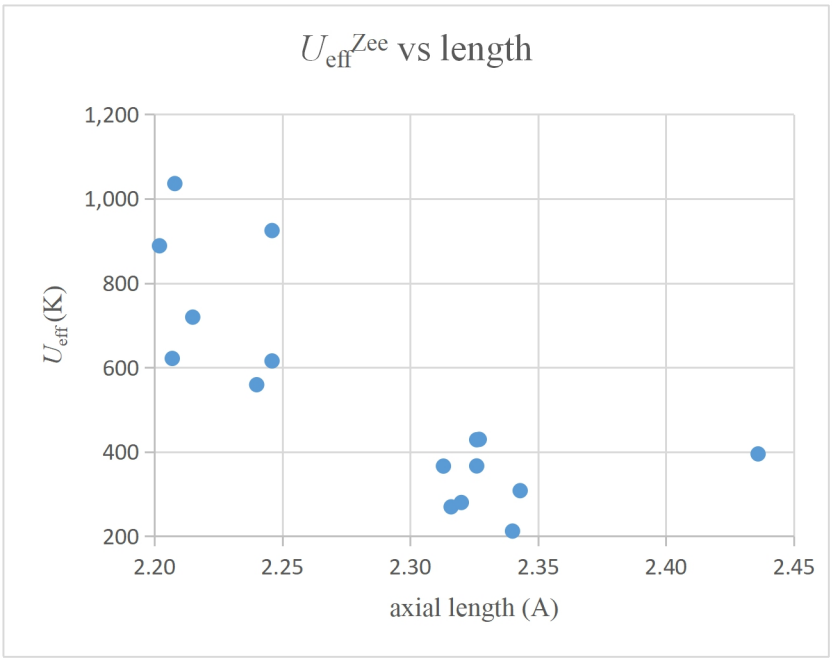

FIG. S18 The relation between U_{eff}^{Zee} and averaged bond length of the two axial ligating atoms

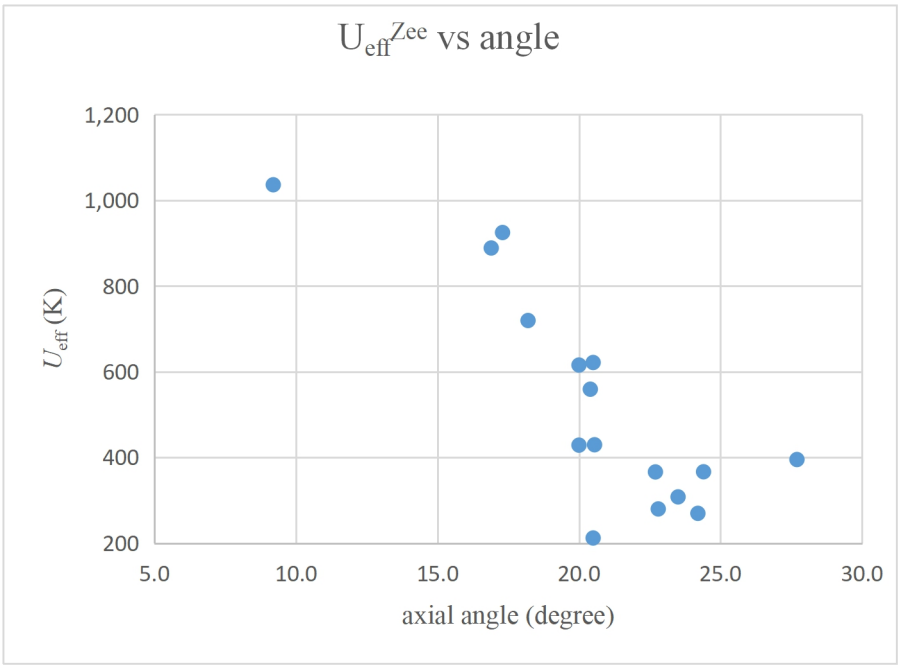

FIG. S19 The relation between U_{eff}^{Zee} and averaged angle of the two axial ligating atoms with respect to magnetic easy axis

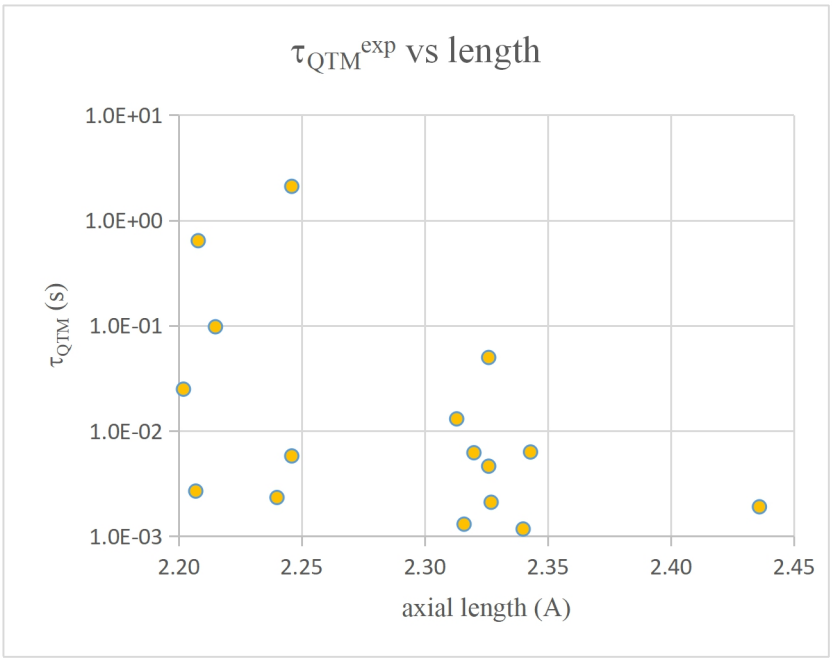

FIG. S20 The relation between τ_{QTM}^{exp} and averaged bond length of the two axial ligating atoms

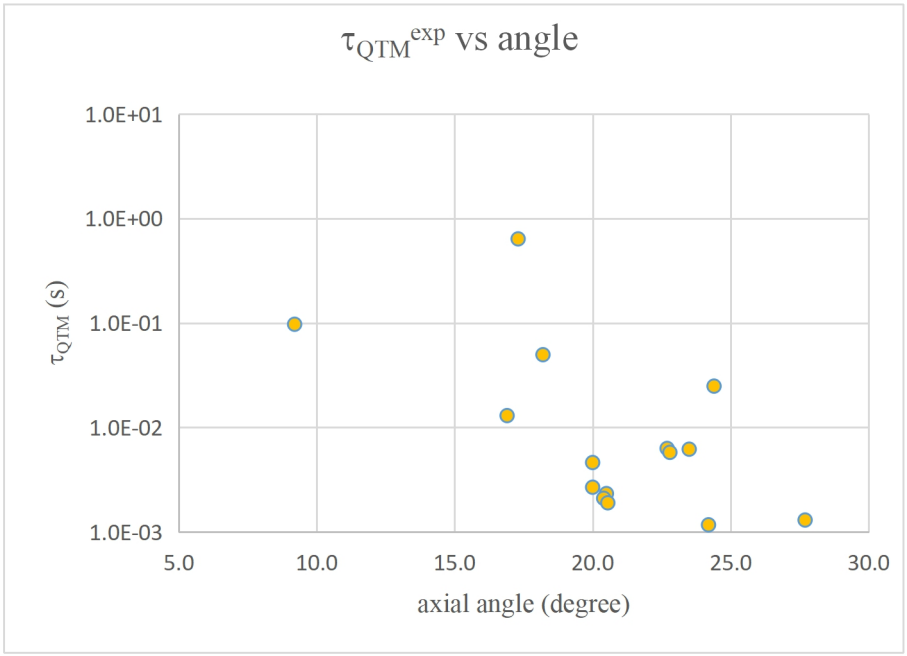

FIG. S21 The relation between τ_{QTM}^{exp} and averaged angle of the two axial ligating atoms with respect to magnetic easy axis

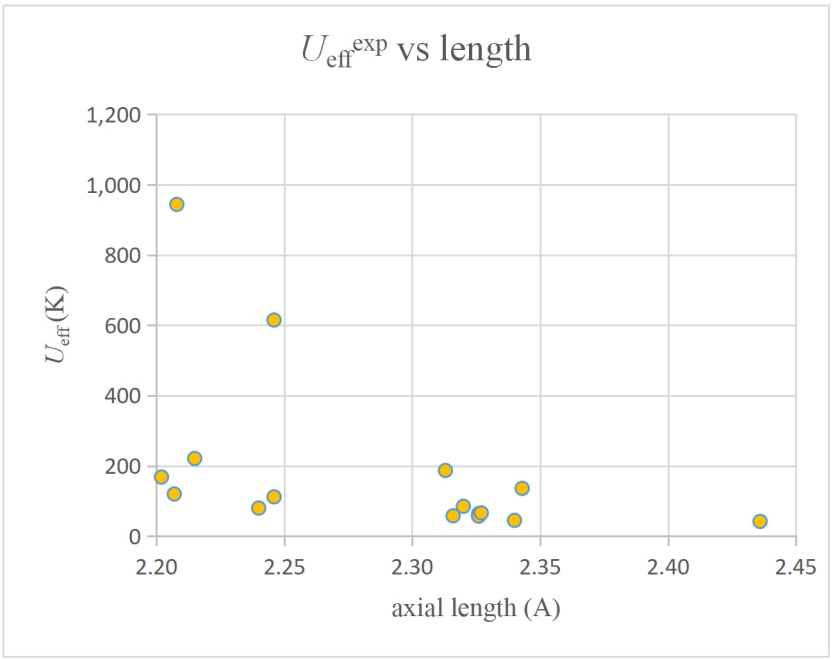

FIG. S22 The relation between U_{eff}^{exp} and averaged bond length of the two axial ligating atoms

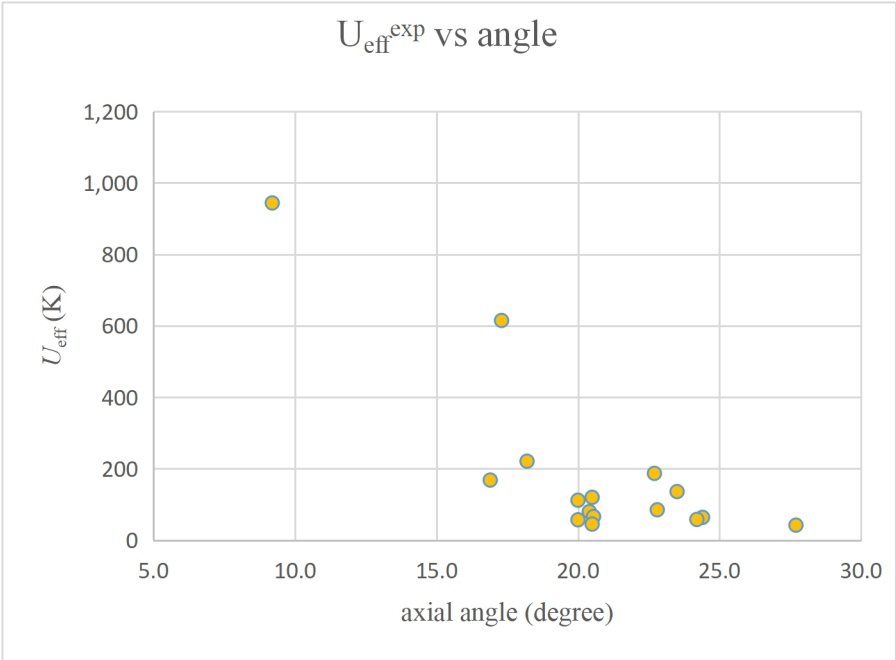

FIG. S23 The relation between U_{eff}^{exp} and averaged angle of the two axial ligating atoms with respect to magnetic easy axis