

### Computational details:

The magnetic exchange interaction between the Gd(III) centers in the non-radical bridged complex has been evaluated using the following Hamiltonian relation,

$$\hat{H} = -2J \hat{S}_{Gd} \hat{S}_{Gd}$$

$J$  represents the Isotropic exchange coupling constant and  $S_1, S_2$  are the spins of Gd (III) centers.  $J$  values are computed from the energy differences between the high spin ( $E_{HS}$ ) state, which is calculated using single determinant wave functions and low spin ( $E_{BS}$ ) state, determined using Broken Symmetry (BS) approach developed by Noddleman.<sup>1, 2</sup> The BS approach was proved to be handy in evaluating the  $J$  values to a good estimate in variety of complexes. All the calculations are performed using hybrid B3LYP<sup>3</sup> functional with a combination of CSDZ ECP on Gd<sup>4</sup> and TZV<sup>5</sup> triple-  $\zeta$  basis set on other atoms as implemented in the Gaussian 09 suite of programs.<sup>6</sup> In the case of radical bridged dinuclear Gd(III) complex, the magnetic interaction between the metal-radical and Gd(III)...Gd(III) interaction has to be taken into account. Four different configurations (all spins up, spin of down on N<sub>2</sub>, spin down on Gd<sub>A</sub>, spin down on Gd<sub>B</sub>) have been computed to obtain the values of  $J_1$  and  $J_2$ .<sup>2</sup>

**Table S1:** DFT computed energies of high spin (HS) and broken symmetry (BS) state,  $\langle S^2 \rangle$  values for different radical systems.

|   | HS          | BS1         | BS2       | BS3       | $\langle S^2 \rangle$ |         |       |       |
|---|-------------|-------------|-----------|-----------|-----------------------|---------|-------|-------|
| <b>1</b>  | -4291.32197 | -4291.32370 | 4291.3229 | 4291.3229 | 63.7782               | 49.7794 | 7.781 | 7.781 |
| <b>2</b>  | -4291.33551 | -4291.33558 | -         | -         | 56.0281               | 7.0295  | -     | -     |
| [Gd(Hbpz <sub>3</sub> ) <sub>2</sub> (dtbsq)](3)                | -2211.03014 | -2211.03036 | -         | -         | 21.0189               | 14.0180 | -     | -     |
| [Gd(NTBzImH) <sub>2</sub> (NO <sub>3</sub> ) <sub>3</sub> ](4)  | -1999.09573 | -1998.92771 | -         | -         | 20.0690               | 13.0578 | -     | -     |
| [Gd(hfac)(IM2py)](5)  | -3640.35909 | -3640.35918 | -         | -         | 20.0284               | 13.0288 | -     | -     |
| [Gd(NTBzImH) <sub>4</sub> ](ClO <sub>4</sub> ) <sub>3</sub> (6) | -1423.30279 | -1423.30284 | -         | -         | 20.0393               | 13.0393 | -     | -     |

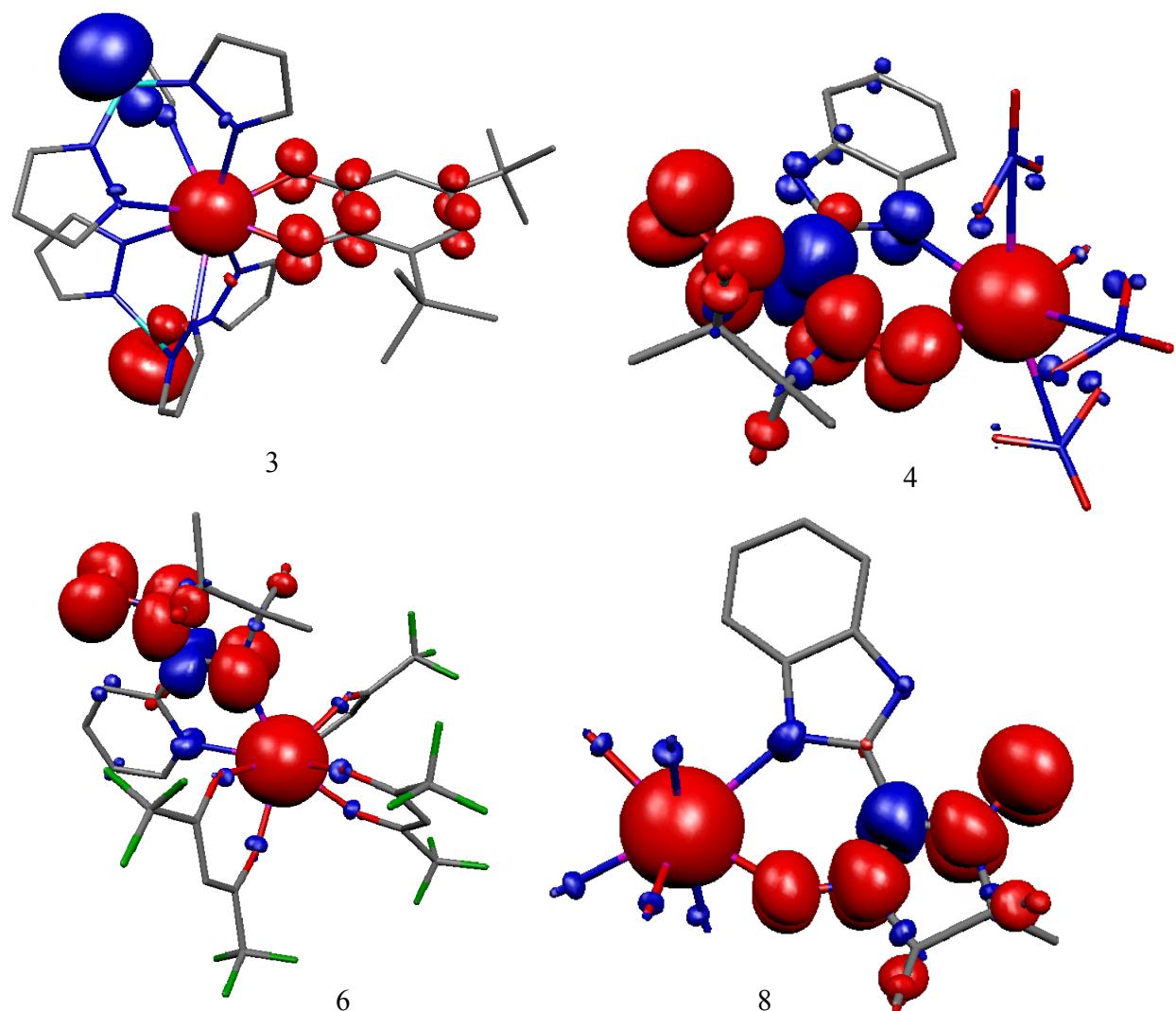
**Table S2:** DFT computed spin densities of selected atoms of complexes **1** and **2**.

| Atom specification | Complex (1)    |         |         |         | Complex (2)    |         |
|--------------------|----------------|---------|---------|---------|----------------|---------|
|                    | Spin densities |         |         |         | Spin densities |         |
|                    | HS             | BS1     | BS2     | BS3     | HS             | BS      |
| Gd1                | 7.0119         | 7.0727  | 7.0337  | -7.0948 | 7.0425         | 7.0571  |
| Gd2                | 7.0152         | 7.0751  | -7.0966 | 7.0364  | 7.0425         | -7.0571 |
| O7                 | 0.0005         | -0.0001 | 0.0003  | 0.0002  | -0.0001        | -0.0003 |

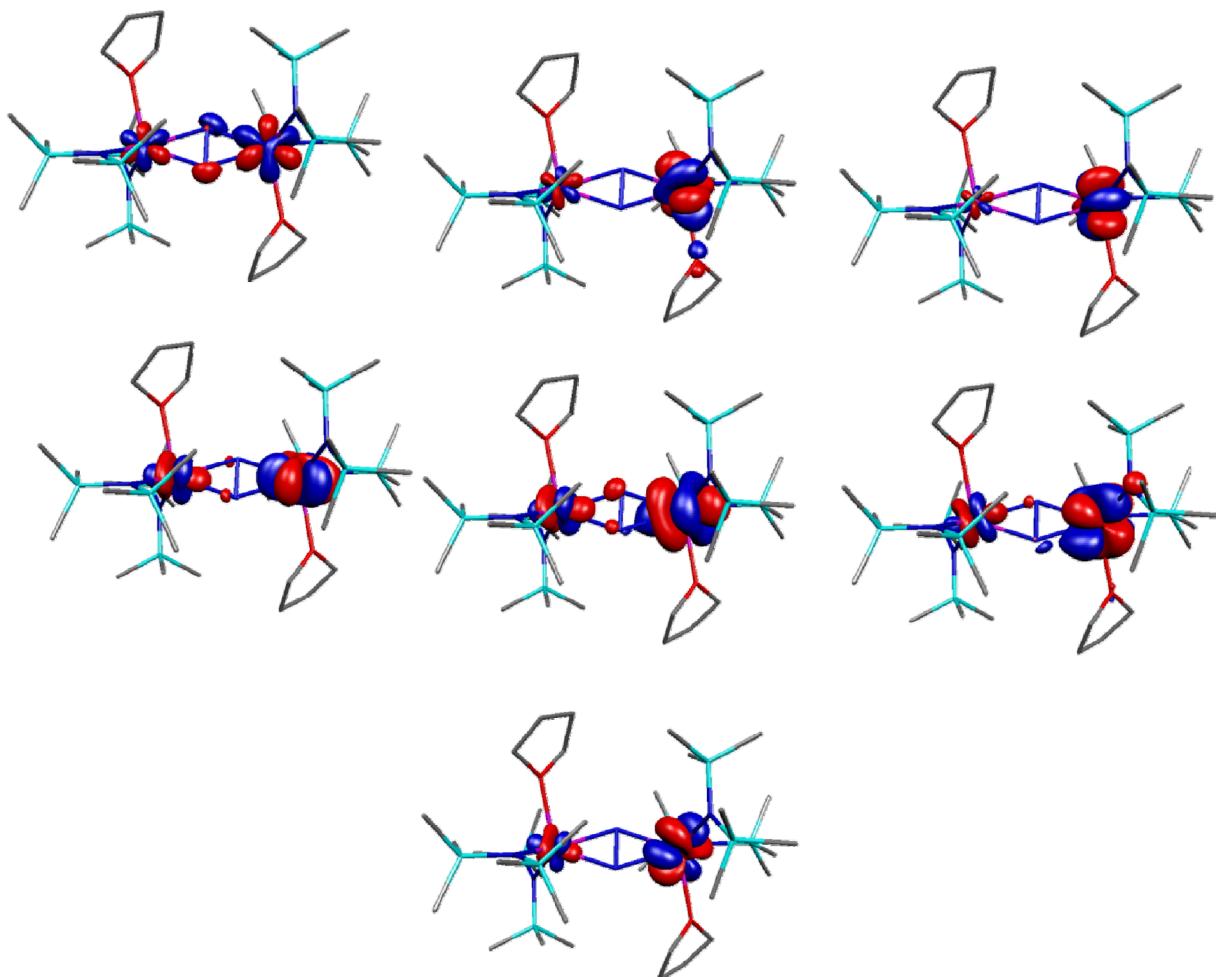
|     |         |         |         |         |         |         |
|-----|---------|---------|---------|---------|---------|---------|
| N8  | -0.0088 | -0.0038 | -0.0087 | 0.0037  | -0.0095 | -0.0095 |
| N9  | -0.0059 | -0.0068 | -0.0058 | 0.0067  | -0.0104 | -0.0104 |
| N10 | 0.4733  | -0.5366 | 0.5021  | 0.5082  | -0.0208 | -0.0011 |
| N71 | 0.4831  | -0.5478 | 0.5208  | 0.5104  | -0.0208 | 0.0011  |
| O76 | 0.0005  | 0.00003 | 0.0001  | 0.0003  | -0.0001 | 0.0003  |
| N77 | -0.0085 | -0.0038 | 0.0037  | -0.0083 | -0.0095 | 0.0095  |
| N78 | -0.0062 | -0.0071 | 0.0070  | -0.0062 | -0.0104 | 0.0104  |

**Table S3:** DFT computed spin densities of selected atoms of complexes **3, 4, 5 and 6**.

| Complex 3              |                |         | Complex 4              |                |         | Complex 5              |                |         | Complex 6              |                |         |
|------------------------|----------------|---------|------------------------|----------------|---------|------------------------|----------------|---------|------------------------|----------------|---------|
| Atom specificati<br>on | Spin densities |         |
|                        | HS             | BS      |
| Gd1                    | 7.0487         | 7.0537  | Gd1                    | 7.0358         | 7.0364  | Gd1                    | 7.0220         | 7.0318  | Gd1                    | 7.0337         | 7.0269  |
| O2                     | 0.2204         | -0.2265 | O2                     | 0.0000         | -0.0002 | O21                    | -0.0013        | -0.0032 | O2                     | 0.1364         | -0.1460 |
| O3                     | 0.1954         | -0.2031 | N4                     | -0.0048        | -0.0044 | O22                    | -0.0019        | -0.0019 | O3                     | 0.4759         | -0.4756 |
| N5                     | 0.0056         | 0.0057  | O7                     | 0.2824         | -0.2858 | O23                    | -0.0019        | -0.0024 | N4                     | 0.2964         | -0.3015 |
| N13                    | -0.0295        | -0.0296 | O8                     | 0.3949         | -0.3941 | O24                    | -0.0023        | -0.0024 | N5                     | 0.2267         | -0.2266 |
| N20                    | -0.0224        | -0.0272 | N9                     | 0.2983         | -0.3006 | O25                    | -0.0005        | -0.0026 | N6                     | -0.0185        | 0.0069  |
| N36                    | 0.0021         | 0.0012  | N10                    | 0.2983         | -0.2978 | O26                    | -0.0010        | -0.0022 | O39                    | -0.0007        | -0.0008 |
| N44                    | 0.0127         | 0.0102  | N11                    | -0.0564        | 0.0472  | N27                    | 0.3251         | -0.3321 | N41                    | -0.0052        | -0.0056 |
|                        |                |         | O45                    | -0.0027        | -0.0041 | N28                    | 0.3163         | -0.3171 | O44                    | -0.0006        | -0.0004 |
|                        |                |         | O46                    | -0.0051        | -0.0050 |                        |                |         | N46                    | -0.0048        | -0.0049 |
|                        |                |         | O49                    | -0.0053        | -0.0057 |                        |                |         | O49                    | -0.0004        | -0.0005 |
|                        |                |         | O50                    | -0.0039        | -0.0037 |                        |                |         | N51                    | -0.0062        | -0.0057 |
|                        |                |         | O53                    | -0.0055        | -0.0076 |                        |                |         |                        |                |         |
|                        |                |         | O54                    | -0.0057        | -0.0066 |                        |                |         |                        |                |         |



**Figure S1:** Computed spin density plot for complexes **3**, **4**, **5** and **6**.



**Figure S2:** Computed magnetic orbitals of the Gd(III) in Complex 1.

Mechanism- Overlap between 4f-orbitals of two Gd(III) ions and  $\pi p_y^*$  of  $N_2^{3-}$

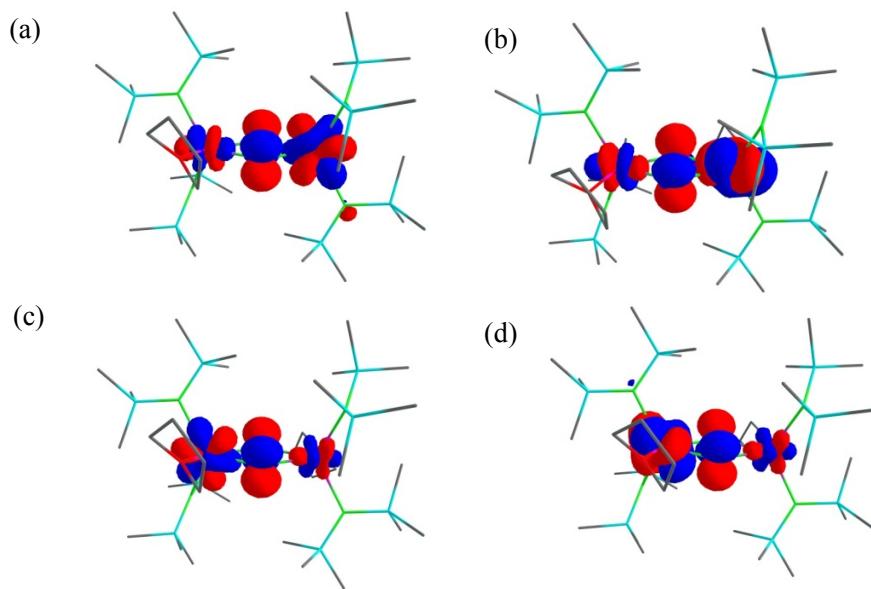
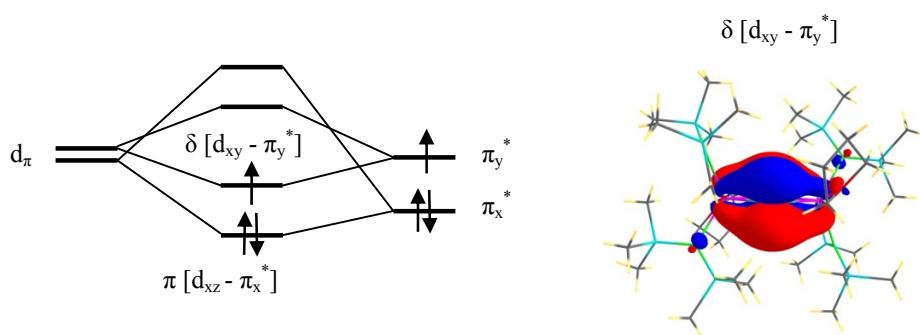


Figure S3. Form overlap integral values (Table S6 and S7), we infer that three 4f orbitals [(a)  $f_{x(x-3y)2}$ , (b and c)  $f_{y(y-2x)2}$ , (d)  $f_{z(x-y)2}$ ] overlap with  $\pi p_y^*$  orbital of  $N_2^{3-}$  plays a prominent role in deciding the interaction as antiferromagnetic and this can be evidenced from the superimposed figure shown above. The overlap of  $f_{y(y-2x)2}$  with  $\pi p_y^*$  orbital is found to be prominent in both Gd(III) ions whereas the  $f_{x(x-3y)2}$  overlap is found to be prominent in Gd<sub>1</sub> and  $f_{z(x-y)2}$  in the case of Gd<sub>2</sub>.



**Figure S3:** Qualitative Molecular diagram for complex 1.

**Table S4:** Exchange coupling values for certain {3d-4f} complexes.

| Complexes   | $J$ (cm <sup>-1</sup> ) | Ref |
|---|-------------------------|-----|
| [Cu <sup>II</sup> Gd <sup>III</sup> {pyCO(OEt)pyC(OH)(OEt)py} <sub>3</sub> ](ClO <sub>4</sub> ) <sub>2</sub> ·EtOH      | -0.32                   | 7   |
| [Mn <sup>II</sup> Gd <sup>III</sup> {pyCO(OEt)pyC(OH)(OEt)py} <sub>3</sub> ](ClO <sub>4</sub> ) <sub>2</sub> ·EtOH      | -1.7                    | 7   |
| [Ni <sup>II</sup> Gd <sup>III</sup> {pyCO(OEt)pyC(OH)(OEt)py} <sub>3</sub> ](ClO <sub>4</sub> ) <sub>2</sub> ·EtOH      | -0.22                   | 7   |
| [LCo(MeOH)Gd(NO <sub>3</sub> ) <sub>3</sub> ]   | 0.45                    | 8   |
| L <sup>1</sup> Fe(CH <sub>3</sub> OH)Gd(NO <sub>3</sub> ) <sub>3</sub> (CH <sub>3</sub> OH) <sub>2</sub>                | 0.50                    | 9   |
| L <sup>1</sup> Fe((CH <sub>3</sub> ) <sub>2</sub> CO)Gd(NO <sub>3</sub> ) <sub>3</sub>                                  | 0.41                    | 9   |
| L <sup>2</sup> Fe((CH <sub>3</sub> ) <sub>2</sub> CO)Gd(NO <sub>3</sub> ) <sub>3</sub>                                  | 0.08                    | 9   |
| [Gd <sub>2</sub> Ni <sub>2</sub> (pro) <sub>4</sub> (NO <sub>3</sub> ) <sub>6</sub> (CH <sub>3</sub> CN) <sub>4</sub> ] | 0.15                    | 10  |
| [LVOGd(hfa) <sub>2</sub> (CH <sub>3</sub> OH)] <sub>2</sub>   | 0.46                    | 11  |
| [L <sup>2</sup> V(O){(CH <sub>3</sub> ) <sub>2</sub> CO}Gd(NO <sub>3</sub> ) <sub>3</sub> ]                             | -2.6                    | 12  |

Magneto-structural correlations (Dihedral Correlation)

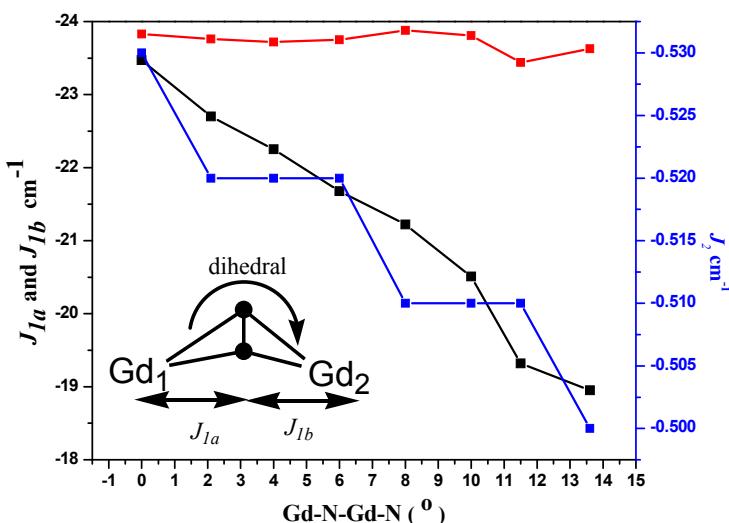


Figure S4. Magneto-structural correlation developed by varying the Gd-N-Gd-N dihedral angle.

The magnetic coupling between  $\text{Gd}_1$  and  $\text{N}_2^{3-}$  has been taken as  $J_{Ia}$  and  $J_{Ib}$  in the case of  $\text{Gd}_2$ . The developed correlation by varying the dihedral angle (Gd-N-Gd-N) is shown in the above figure, in which  $J_{Ia}$  is denoted in red lines and  $J_{Ib}$  in black lines whereas  $J_2$  (interaction between 2 Gd(III) atoms) is denoted by blue lines. Here the dihedral is varied by moving the  $\text{Gd}_2$  atom out of the  $\text{Gd}_2\text{N}_2$  plane without affecting the position of  $\text{Gd}_1$ . Upon varying the dihedral,  $J_{Ia}$  remains unaffected whereas  $J_{Ib}$  and  $J_2$  vary constantly. This variation routes from the large difference between the overlap between the 4f-orbitals and  $\pi\text{p}_y^*$  (Table S7 and S9).

**Table S5:** Spin densities and NPA charges of different atoms in complex 1.

| Distance (N-N) | N1    | N2    |
|----------------|-------|-------|
| 1.440          | 0.488 | 0.498 |
| 1.420          | 0.480 | 0.490 |
| 1.400          | 0.473 | 0.483 |
| 1.380          | 0.466 | 0.474 |
| 1.360          | 0.456 | 0.465 |
| 1.340          | 0.446 | 0.455 |
| 1.320          | 0.434 | 0.443 |

|       |       |       |
|-------|-------|-------|
| 1.300 | 0.421 | 0.431 |
| 1.278 | 0.409 | 0.417 |
| 1.260 | 0.395 | 0.402 |
| 1.240 | 0.378 | 0.385 |

| Distance<br>(N-N)  | Gd1   | Gd2   | N1     | N2     |
|--------------------|-------|-------|--------|--------|
| 1.400 ( <b>1</b> ) | 1.743 | 1.700 | -0.813 | -0.793 |
| 1.380              | 1.734 | 1.689 | -0.806 | -0.788 |
| 1.320              | 1.693 | 1.640 | -0.777 | -0.759 |
| 1.278              | 1.665 | 1.600 | -0.748 | -0.731 |
| 1.240              | 1.636 | 1.570 | -0.717 | -0.701 |

**Table S6:** Overlap integral ( $S_{ab}$ ) values for distance correlation in Complex 1 corresponding to coupling constant  $J_1$  ( $J_{la}$  and  $J_{lb}$ ).

|                |                   |                 |             |             |                   |                 |              |             |                   |                 |              |              |                   |                 |              |
|----------------|-------------------|-----------------|-------------|-------------|-------------------|-----------------|--------------|-------------|-------------------|-----------------|--------------|--------------|-------------------|-----------------|--------------|
| <b>1.4</b>     | $f_{xz^2}$        | $\pi p_y^*$     | -0.01664    | <b>1.36</b> | $f_{xz^2}$        | $\pi p_y^*$     | 0.02371      | <b>1.32</b> | $f_{xz^2}$        | $\pi p_y^*$     | 0.00315      | <b>1.278</b> | $f_{xz^2}$        | $\pi p_y^*$     | 0.02413      |
| <b>Gd1</b>     | $f_{yz^2}$        |                 | -0.01925    |             | $f_{x(x^2-3y^2)}$ |                 | 0.04579      |             |                   |                 | -0.03830     |              | $f_{xyz}$         |                 | -0.02868     |
|                | $f_{z^3}$         |                 | 0.01346     |             | $f_{z^3}$         |                 | 0.01180      |             | $f_{z^3}$         |                 | 0.01175      |              | $f_{z^3}$         |                 | -0.00531     |
|                | $f_{z(x^2-y^2)}$  |                 | 0.01493     |             | $f_{yz^2}$        |                 | 0.00970      |             | $f_{yz^2}$        |                 | 0.00575      |              | $f_{yz^2}$        |                 | 0.00249      |
|                | $f_{xxyz}$        |                 | 0.00613     |             | $f_{y(y^2-3x^2)}$ |                 | 0.04274      |             |                   |                 | 0.04773      |              | $f_{x(x^2-3y^2)}$ |                 | 0.03399      |
|                | $f_{x(x^2-3y^2)}$ |                 | 0.05232     |             | $f_{xyz}$         |                 | 0.00481      |             | $f_{xyz}$         |                 | 0.00722      |              | $f_{y(y^2-3x^2)}$ |                 | -0.03548     |
|                | $f_{y(y^2-3x^2)}$ |                 | 0.03692     |             | $f_{z(x^2-y^2)}$  |                 | 0.01575      |             |                   |                 | 0.01425      |              | $f_{z(x^2-y^2)}$  |                 | 0.02320      |
| <br><b>Gd2</b> | <br>$f_{xz^2}$    | <br>$\pi p_y^*$ | <br>0.00995 |             | <br>$f_{xz^2}$    | <br>$\pi p_y^*$ | <br>-0.01451 |             | <br>$f_{xz^2}$    | <br>$\pi p_y^*$ | <br>-0.01362 |              | <br>$f_{xz^2}$    | <br>$\pi p_y^*$ | <br>-0.00673 |
|                | $f_{yz^2}$        |                 | 0.01836     |             | $f_{xyz}$         |                 | 0.01119      |             | $f_{xyz}$         |                 | -0.00591     |              | $f_{xyz}$         |                 | -0.00041     |
|                | $f_{z^3}$         |                 | -0.00817    |             | $f_{z^3}$         |                 | -0.01063     |             | $f_{z^3}$         |                 | -0.01279     |              | $f_{z^3}$         |                 | -0.00230     |
|                | $f_{z(x^2-y^2)}$  |                 | 0.02466     |             | $f_{yz^2}$        |                 | -0.00942     |             | $f_{yz^2}$        |                 | -0.00242     |              | $f_{yz^2}$        |                 | 0.00127      |
|                | $f_{x(x^2-3y^2)}$ |                 | 0.01603     |             | $f_{y(y^2-3x^2)}$ |                 | -0.06184     |             | $f_{y(y^2-3x^2)}$ |                 | -0.06682     |              | $f_{y(y^2-3x^2)}$ |                 | -0.06964     |
|                | $f_{xyz}$         |                 | -0.01277    |             | $f_{z(x^2-y^2)}$  |                 | -0.02493     |             | $f_{z(x^2-y^2)}$  |                 | -0.02275     |              | $f_{z(x^2-y^2)}$  |                 | 0.00302      |
|                | $f_{y(y^2-3x^2)}$ |                 | -0.0634     |             | $f_{x(x^2-3y^2)}$ |                 | 0.01069      |             |                   |                 | -0.00055     |              | $f_{x(x^2-3y^2)}$ |                 | -0.00601     |

**Table S7:** Overlap integral ( $S_{ab}$ ) values for dihedral correlation in Complex 1 corresponding to coupling constant  $J_I$  ( $J_{la}$  and  $J_{lb}$ )

|                |                   |                 |             |          |                   |             |          |           |                   |             |          |             |                   |             |          |
|----------------|-------------------|-----------------|-------------|----------|-------------------|-------------|----------|-----------|-------------------|-------------|----------|-------------|-------------------|-------------|----------|
| <b>0</b>       | $f_{xz^2}$        | $\pi p_y^*$     | -0.01664    | <b>4</b> | $f_{xz^2}$        | $\pi p_y^*$ | -0.01264 | <b>10</b> | $f_{xz^2}$        | $\pi p_y^*$ | -0.00964 | <b>13.6</b> | $f_{xz^2}$        | $\pi p_y^*$ | -0.01178 |
| <b>Gd1</b>     | $f_{yz^2}$        |                 | -0.01925    |          | $f_{yz^2}$        |             | -0.0056  |           | $f_{yz^2}$        |             | 0.002887 |             | $f_{yz^2}$        |             | 0.022082 |
|                | $f_z^3$           |                 | 0.01346     |          | $f_z^3$           |             | -0.01433 |           | $f_z^3$           |             | -0.01559 |             | $f_z^3$           |             | -0.01478 |
|                | $f_{z(x^2-y^2)}$  |                 | 0.01493     |          | $f_{xyz}$         |             | -0.01973 |           | $f_{xyz}$         |             | -0.02076 |             | $f_{xyz}$         |             | -0.02019 |
|                | $f_{xyz}$         |                 | 0.00613     |          | $f_{(y^2-3x^2)}$  |             | 0.050831 |           | $f_{(y^2-3x^2)}$  |             | 0.047755 |             | $f_{(y^2-3x^2)}$  |             | -0.04391 |
|                | $f_{x(x^2-3y^2)}$ |                 | 0.05232     |          | $f_{x(x^2-3y^2)}$ |             | -0.03949 |           | $f_{x(x^2-3y^2)}$ |             | 0.044404 |             | $f_{x(x^2-3y^2)}$ |             | 0.044373 |
|                | $f_{y(y^2-3x^2)}$ |                 | 0.03692     |          | $f_{z(x^2-y^2)}$  |             | -0.01566 |           | $f_{z(x^2-y^2)}$  |             | -0.01629 |             | $f_{z(x^2-y^2)}$  |             | -0.01553 |
| <br><b>Gd2</b> | <br>$f_{xz^2}$    | <br>$\pi p_y^*$ | <br>0.00995 |          | $f_{xz^2}$        | $\pi p_y^*$ | 0.015593 |           | $f_{xz^2}$        | $\pi p_y^*$ | 0.024332 |             | $f_{xz^2}$        | $\pi p_y^*$ | 0.030052 |
|                | $f_{yz^2}$        |                 | 0.01836     |          | $f_{yz^2}$        |             | 0.009635 |           | $f_{yz^2}$        |             | 0.006456 |             | $f_{yz^2}$        |             | -0.00057 |
|                | $f_z^3$           |                 | -0.00817    |          | $f_z^3$           |             | 0.00628  |           | $f_z^3$           |             | 0.005635 |             | $f_z^3$           |             | 0.004117 |
|                | $f_{z(x^2-y^2)}$  |                 | 0.02466     |          | $f_{(x^2-3y^2)}$  |             | 0.017923 |           | $f_{(x^2-3y^2)}$  |             | 0.01611  |             | $f_{(x^2-3y^2)}$  |             | 0.013074 |
|                | $f_{x(x^2-3y^2)}$ |                 | 0.01603     |          | $f_{(y^2-3x^2)}$  |             | -0.06267 |           | $f_{(y^2-3x^2)}$  |             | -0.06055 |             | $f_{(y^2-3x^2)}$  |             | -0.05986 |
|                | $f_{xyz}$         |                 | -0.01277    |          | $f_{z(x^2-y^2)}$  |             | -0.02729 |           | $f_{z(x^2-y^2)}$  |             | -0.02937 |             | $f_{z(x^2-y^2)}$  |             | -0.02653 |
|                | $f_{y(y^2-3x^2)}$ |                 | -0.0634     |          | $f_{xyz}$         |             | 0.012418 |           | $f_{xyz}$         |             | 0.002866 |             | $f_{xyz}$         |             | -0.00345 |

**Table S8:** Overlap integral ( $S_{ab}$ ) values for distance correlation in complex 1 corresponding to coupling constant  $J_2$ .

|            |                   |                 |             |                   |                 |             |                   |                |              |            |                   |                 |
|------------|-------------------|-----------------|-------------|-------------------|-----------------|-------------|-------------------|----------------|--------------|------------|-------------------|-----------------|
| <b>1.4</b> | $f_{xz^2}$        | <b>-0.01465</b> | <b>1.36</b> | $f_{xz^2}$        | <b>0.005901</b> | <b>1.32</b> | $f_{xz^2}$        | <b>0.00256</b> | <b>1.278</b> | $f_{xz^2}$ | $f_{xz^2}$        | <b>0.001416</b> |
|            | $f_{yz^2}$        | -0.000359       |             | $f_{yz^2}$        | 0.000525        |             | $f_{yz^2}$        | -0.000188      |              |            | $f_{yz^2}$        | -0.00047        |
|            | $f_z^3$           | 0.0029013       |             | $f_z^3$           | -0.005179       |             | $f_z^3$           | -0.00339       |              |            | $f_z^3$           | -0.00188        |
|            | $f_{z(x^2-y^2)}$  | 0.0076368       |             | $f_{z(x^2-y^2)}$  | -0.00417        |             | $f_{z(x^2-y^2)}$  | -0.001098      |              |            | $f_{z(x^2-y^2)}$  | -0.000496       |
|            | $f_{xyz}$         | -0.000112       |             | $f_{xyz}$         | 0.0002087       |             | $f_{xyz}$         | 0.0001526      |              |            | $f_{xyz}$         | 6.818E-05       |
|            | $f_{y(y^2-3x^2)}$ | 0.0014042       |             | $f_{y(y^2-3x^2)}$ | 0.0020709       |             | $f_{y(y^2-3x^2)}$ | 0.0015879      |              |            | $f_{y(y^2-3x^2)}$ | 0.0011658       |
|            | $f_{x(x^2-3y^2)}$ | 0.0003957       |             | $f_{x(x^2-3y^2)}$ | 0.0000511       |             | $f_{x(x^2-3y^2)}$ | 0.0005359      |              |            | $f_{x(x^2-3y^2)}$ | 0.0005559       |
|            | $f_{y^2}$         | 0.0076368       |             | $f_{y^2}$         | $f_{xz^2}$      | -0.003104   | $f_{xz^2}$        | -0.001476      | $f_{xz^2}$   | $f_{xz^2}$ | -0.000714         |                 |
|            | $f_{yz^2}$        | -0.000724       |             | $f_{yz^2}$        | -0.000997       |             | $f_{yz^2}$        | -0.00042       |              |            | $f_{yz^2}$        | -0.000186       |
|            | $f_z^3$           | -0.001588       |             | $f_z^3$           | 0.0030758       |             | $f_z^3$           | 0.0023283      |              |            | $f_z^3$           | 0.0012764       |
|            | $f_{z(x^2-y^2)}$  | -0.002268       |             | $f_{z(x^2-y^2)}$  | 0.0024185       |             | $f_{z(x^2-y^2)}$  | 0.0006721      |              |            | $f_{z(x^2-y^2)}$  | -0.000269       |
|            | $f_{xyz}$         | -0.000464       |             | $f_{xyz}$         | 0.0002817       |             | $f_{xyz}$         | 9.294E-05      |              |            | $f_{xyz}$         | 7.693E-05       |
|            | $f_{y(y^2-3x^2)}$ | -0.001084       |             | $f_{y(y^2-3x^2)}$ | -0.00084        |             | $f_{y(y^2-3x^2)}$ | -0.000945      |              |            | $f_{y(y^2-3x^2)}$ | -0.00075        |
|            | $f_{x(x^2-3y^2)}$ | 0.0001319       |             | $f_{x(x^2-3y^2)}$ | -5.95E-05       |             | $f_{x(x^2-3y^2)}$ | -0.000378      |              |            | $f_{x(x^2-3y^2)}$ | -0.000398       |
|            | $f_z^3$           | 0.0029013       |             | $f_z^3$           | $f_{xz^2}$      | -0.002654   | $f_z^3$           | $f_{xz^2}$     | -0.002559    |            | $f_z^3$           | -0.002057       |
|            | $f_{yz^2}$        | 0.0009341       |             | $f_{yz^2}$        | -0.000828       |             | $f_{yz^2}$        | -0.000569      |              |            | $f_{yz^2}$        | -3.58E-05       |
|            | $f_z^3$           | 0.0007919       |             | $f_z^3$           | 0.0022352       |             | $f_z^3$           | 0.0034652      |              |            | $f_z^3$           | 0.002802        |
|            | $f_{z(x^2-y^2)}$  | -0.001588       |             | $f_{z(x^2-y^2)}$  | 0.0020252       |             | $f_{z(x^2-y^2)}$  | 0.0012885      |              |            | $f_{z(x^2-y^2)}$  | 0.0009067       |
|            | $f_{xyz}$         | 0.0005178       |             | $f_{xyz}$         | 0.0003188       |             | $f_{xyz}$         | 0.000138       |              |            | $f_{xyz}$         | 2.769E-05       |
|            | $f_{y(y^2-3x^2)}$ | -0.0005932      |             | $f_{y(y^2-3x^2)}$ | -0.000412       |             | $f_{y(y^2-3x^2)}$ | -0.001208      |              |            | $f_{y(y^2-3x^2)}$ | -0.001506       |
|            | $f_{x(x^2-3y^2)}$ | 0.0001651       |             | $f_{x(x^2-3y^2)}$ | 0.0000319       |             | $f_{x(x^2-3y^2)}$ | -0.000444      |              |            | $f_{x(x^2-3y^2)}$ | -0.00068        |

|                   |            |                   |            |                  |                   |            |                   |            |
|-------------------|------------|-------------------|------------|------------------|-------------------|------------|-------------------|------------|
| $f_{z(x^2-y^2)}$  | $f_{xz^2}$ | $f_{yz^2}$        | $f_{zx^2}$ | $f_{xy^2}$       | $f_{xz^2}$        | $f_{yz^2}$ | $f_{zx^2}$        | $f_{xy^2}$ |
| $f_{xz^2}$        | 0.0003957  | $f_{x^2}$         | -0.000295  | $f_{y^2}$        | $f_{x^2}$         | -5.86E-05  | $f_{y^2}$         | 7.014E-05  |
| $f_{yz^2}$        | 0.0001282  | $f_{y^2}$         | 0.0001167  | $f_{x^2}$        | $f_{yz^2}$        | 0.0003092  | $f_{x^2}$         | 0.000512   |
| $f_z$             | 0.0001651  | $f_z$             | -0.000146  | $f_z$            | $f_z$             | -0.00027   | $f_z$             | -0.000344  |
| $f_{z(x^2-y^2)}$  | 0.0001319  | $f_{z(x^2-y^2)}$  | -0.000085  | $f_{z(x^2-y^2)}$ | $f_{z(x^2-y^2)}$  | -0.000127  | $f_{z(x^2-y^2)}$  | -0.000218  |
| $f_{xyz}$         | -0.000889  | $f_{xyz}$         | -0.000779  | $f_{xyz}$        | $f_{xyz}$         | -0.000534  | $f_{xyz}$         | -0.00034   |
| $f_{(y^2-3x^2)}$  | 0.0003516  | $f_{(y^2-3x^2)}$  | -0.000864  | $f_{(y^2-3x^2)}$ | $f_{(y^2-3x^2)}$  | -0.000788  | $f_{(y^2-3x^2)}$  | -0.000547  |
| $f_{(x^2-3y^2)}$  | 0.0013567  | $f_{(x^2-3y^2)}$  | 0.0011173  | $f_{(x^2-3y^2)}$ | $f_{(x^2-3y^2)}$  | 0.0008628  | $f_{(x^2-3y^2)}$  | 0.0008658  |
| $f_{x(x^2-3y^2)}$ | $f_{xz^2}$ | -0.000359         | $f_{xz^2}$ | -0.000512        | $f_{xz^2}$        | -0.000522  | $f_{xz^2}$        | -0.000468  |
| $f_{y^2}$         | 0.0012951  | $f_{y^2}$         | $f_{yz^2}$ | 0.0024302        | $f_{yz^2}$        | 0.0027463  | $f_{yz^2}$        | 0.0029845  |
| $f_z$             | 0.0009341  | $f_z$             | $f_z$      | -0.000917        | $f_z$             | -0.000682  | $f_z$             | -0.00056   |
| $f_{z(x^2-y^2)}$  | -0.000724  | $f_{z(x^2-y^2)}$  | -0.000333  | $f_{z(x^2-y^2)}$ | $f_{z(x^2-y^2)}$  | -6.04E-05  | $f_{z(x^2-y^2)}$  | -0.000203  |
| $f_{xyz}$         | 0.0001596  | $f_{xyz}$         | -0.001513  | $f_{xyz}$        | $f_{xyz}$         | -0.00129   | $f_{xyz}$         | -0.000984  |
| $f_{(y^2-3x^2)}$  | -0.001558  | $f_{(y^2-3x^2)}$  | -0.000346  | $f_{(y^2-3x^2)}$ | $f_{(y^2-3x^2)}$  | -0.000276  | $f_{(y^2-3x^2)}$  | 5.044E-05  |
| $f_{(x^2-3y^2)}$  | 0.0001282  | $f_{(x^2-3y^2)}$  | 0.0000017  | $f_{(x^2-3y^2)}$ | $f_{(x^2-3y^2)}$  | 9.336E-05  | $f_{(x^2-3y^2)}$  | 0.0001434  |
| $f_{xyz}$         | 0.0014042  | $f_{xyz}$         | $f_{xz^2}$ | 0.0009255        | $f_{xyz}$         | 0.0007726  | $f_{xyz}$         | 0.0006775  |
| $f_{yz^2}$        | -0.001558  | $f_{yz^2}$        | $f_{yz^2}$ | 0.0012591        | $f_{yz^2}$        | 0.001057   | $f_{yz^2}$        | 0.0007042  |
| $f_z$             | -0.0005932 | $f_z$             | $f_z$      | -0.00099         | $f_z$             | -0.001207  | $f_z$             | -0.000993  |
| $f_{z(x^2-y^2)}$  | -0.001084  | $f_{z(x^2-y^2)}$  | -0.000938  | $f_{z(x^2-y^2)}$ | $f_{z(x^2-y^2)}$  | -0.000556  | $f_{z(x^2-y^2)}$  | -0.00046   |
| $f_{xyz}$         | 0.0008753  | $f_{xyz}$         | -0.00056   | $f_{xyz}$        | $f_{xyz}$         | -0.000307  | $f_{xyz}$         | -0.000117  |
| $f_{(y^2-3x^2)}$  | -0.001211  | $f_{(y^2-3x^2)}$  | -0.000156  | $f_{(y^2-3x^2)}$ | $f_{(y^2-3x^2)}$  | 0.0002236  | $f_{(y^2-3x^2)}$  | 0.0004589  |
| $f_{(x^2-3y^2)}$  | 0.0003516  | $f_{(x^2-3y^2)}$  | -0.000164  | $f_{(x^2-3y^2)}$ | $f_{(x^2-3y^2)}$  | -5.61E-05  | $f_{(x^2-3y^2)}$  | 5.012E-05  |
| $f_{y(y^2-3x^2)}$ | -0.000112  | $f_{y(y^2-3x^2)}$ | $f_{xz^2}$ | 0.0007226        | $f_{y(y^2-3x^2)}$ | 0.0005271  | $f_{y(y^2-3x^2)}$ | 0.0004642  |

|                   |           |                   |           |                   |           |                   |           |
|-------------------|-----------|-------------------|-----------|-------------------|-----------|-------------------|-----------|
| $f_{yz^2}$        | 0.0001596 | $f_{yz^2}$        | -0.001038 | $f_{yz^2}$        | -0.00097  | $f_{yz^2}$        | -0.000865 |
| $f_z^3$           | 0.0005178 | $f_z^3$           | 0.0003537 | $f_z^3$           | 1.964E-05 | $f_z^3$           | -0.000156 |
| $f_{z(x^2-y^2)}$  | -0.000464 | $f_{z(x^2-y^2)}$  | -9.62E-05 | $f_{z(x^2-y^2)}$  | -0.000161 | $f_{z(x^2-y^2)}$  | -0.000107 |
| $f_{xyz}$         | 0.0011235 | $f_{xyz}$         | 0.0007971 | $f_{xyz}$         | 0.0004723 | $f_{xyz}$         | 0.0002081 |
| $f_{y(y^2-3x^2)}$ | 0.0008753 | $f_{y(y^2-3x^2)}$ | 0.0007173 | $f_{y(y^2-3x^2)}$ | 0.0006543 | $f_{y(y^2-3x^2)}$ | 0.0004825 |
| $f_{x(x^2-3y^2)}$ | -0.000889 | $f_{x(x^2-3y^2)}$ | -0.000753 | $f_{x(x^2-3y^2)}$ | -0.000551 | $f_{x(x^2-3y^2)}$ | -0.000428 |

**Table S9:** Overlap integral ( $S_{ab}$ ) values for dihedral correlation in Complex 1 corresponding to coupling constant  $J_2$ .

|          |                   |                   |                 |          |                   |                 |           |                   |                 |             |                   |                   |                 |
|----------|-------------------|-------------------|-----------------|----------|-------------------|-----------------|-----------|-------------------|-----------------|-------------|-------------------|-------------------|-----------------|
| <b>0</b> | $f_{xz^2}$        | $f_{xz^2}$        | <b>-0.01465</b> | <b>4</b> | $f_{xz^2}$        | <b>-0.01401</b> | <b>10</b> | $f_{xz^2}$        | <b>-0.01301</b> | <b>13.6</b> | $f_{xz^2}$        | $f_{xz^2}$        | <b>-0.01043</b> |
|          | $f_{yz^2}$        | $f_{yz^2}$        | -0.000359       |          | $f_{yz^2}$        | 0.002171        |           | $f_{yz^2}$        | -0.00102        |             | $f_{xz^2}$        | $f_{xz^2}$        | -0.00177        |
|          | $f_z^3$           | $f_z^3$           | 0.0029013       |          | $f_z^3$           | 0.006629        |           | $f_z^3$           | 0.00912         |             | $f_z^3$           | $f_z^3$           | 0.01154         |
|          | $f_{z(x^2-y^2)}$  | $f_{z(x^2-y^2)}$  | 0.0076368       |          | $f_{z(x^2-y^2)}$  | 0.007243        |           | $f_{z(x^2-y^2)}$  | 0.006789        |             | $f_{yz^2}$        | $f_{yz^2}$        | 0.005636        |
|          | $f_{xyz}$         | $f_{xyz}$         | -0.000112       |          | $f_{xyz}$         | -0.00045        |           | $f_{xyz}$         | -0.0011         |             | $f_{y(y^2-3x^2)}$ | $f_{y(y^2-3x^2)}$ | -0.00154        |
|          | $f_{y(y^2-3x^2)}$ | $f_{y(y^2-3x^2)}$ | 0.0014042       |          | $f_{y(y^2-3x^2)}$ | -0.00138        |           | $f_{y(y^2-3x^2)}$ | 0.002411        |             | $f_{x(x^2-3y^2)}$ | $f_{x(x^2-3y^2)}$ | 0.00237         |
|          | $f_{x(x^2-3y^2)}$ | $f_{x(x^2-3y^2)}$ | 0.0003957       |          | $f_{x(x^2-3y^2)}$ | 0.00062         |           | $f_{x(x^2-3y^2)}$ | 0.001479        |             | $f_{xyz}$         | $f_{xyz}$         | 0.001379        |
|          | $f_{yz^2}$        | $f_{yz^2}$        | 0.0076368       |          | $f_{yz^2}$        | -0.00364        |           | $f_{yz^2}$        | -0.00333        |             | $f_{yz^2}$        | $f_{yz^2}$        | -0.00327        |
|          | $f_{yz^2}$        | $f_{yz^2}$        | -0.000724       |          | $f_{yz^2}$        | 0.000809        |           | $f_{yz^2}$        | 0.000588        |             | $f_{xz^2}$        | $f_{xz^2}$        | 0.000414        |
|          | $f_z^3$           | $f_z^3$           | -0.001588       |          | $f_z^3$           | 0.002006        |           | $f_z^3$           | 0.002696        |             | $f_z^3$           | $f_z^3$           | 0.004029        |
|          | $f_{z(x^2-y^2)}$  | $f_{z(x^2-y^2)}$  | -0.002268       |          | $f_{z(x^2-y^2)}$  | 0.002119        |           | $f_{z(x^2-y^2)}$  | 0.001903        |             | $f_{yz^2}$        | $f_{yz^2}$        | 0.001858        |
|          | $f_{xyz}$         | $f_{xyz}$         | -0.000464       |          | $f_{xyz}$         | -0.00065        |           | $f_{xyz}$         | -0.00083        |             | $f_{y(y^2-3x^2)}$ | $f_{y(y^2-3x^2)}$ | -0.00113        |
|          | $f_{y(y^2-3x^2)}$ | $f_{y(y^2-3x^2)}$ | -0.001084       |          | $f_{y(y^2-3x^2)}$ | 0.000422        |           | $f_{y(y^2-3x^2)}$ | 0.000453        |             | $f_{x(x^2-3y^2)}$ | $f_{x(x^2-3y^2)}$ | 0.000501        |
|          | $f_{x(x^2-3y^2)}$ | $f_{x(x^2-3y^2)}$ | 0.0001319       |          | $f_{x(x^2-3y^2)}$ | 0.000244        |           | $f_{x(x^2-3y^2)}$ | 0.000506        |             | $f_{xyz}$         | $f_{xyz}$         | 0.000645        |

|                  |            |           |                  |            |          |                  |            |          |                  |            |          |
|------------------|------------|-----------|------------------|------------|----------|------------------|------------|----------|------------------|------------|----------|
| $f_z$            | $f_{xz^2}$ | 0.0029013 | $f_z$            | $f_{xz^2}$ | -0.00279 | $f_z$            | $f_{xz^2}$ | -0.00258 | $f_z$            | $f_{xz^2}$ | -0.00277 |
| $f_{yz^2}$       | 0.0009341  |           | $f_{yz^2}$       | 0.000392   |          | $f_{yz^2}$       | 0.000501   |          | $f_{yz^2}$       | 0.000433   |          |
| $f_z$            | 0.0007919  |           | $f_z$            | 0.001137   |          | $f_z$            | 0.001648   |          | $f_z$            | 0.003017   |          |
| $f_{z(x^2-y^2)}$ | -0.001588  |           | $f_{z(x^2-y^2)}$ | 0.00153    |          | $f_{z(x^2-y^2)}$ | 0.001432   |          | $f_{z(x^2-y^2)}$ | 0.001602   |          |
| $f_{xyz}$        | 0.0005178  |           | $f_{xyz}$        | -0.00056   |          | $f_{xyz}$        | -0.00065   |          | $f_{xyz}$        | -0.00096   |          |
| $f_{(y^2-3x^2)}$ | 0.0005932  | -         | $f_{(y^2-3x^2)}$ | 0.00047    |          | $f_{(y^2-3x^2)}$ | 0.000116   |          | $f_{(y^2-3x^2)}$ | 0.000126   |          |
| $f_{(x^2-3y^2)}$ | 0.0001651  |           | $f_{(x^2-3y^2)}$ | 0.000258   |          | $f_{(x^2-3y^2)}$ | 0.000427   |          | $f_{(x^2-3y^2)}$ | 0.000516   |          |
| $f_{z(x^2-y^2)}$ |            |           | $f_{z(x^2-y^2)}$ |            |          | $f_{z(x^2-y^2)}$ |            |          | $f_{z(x^2-y^2)}$ |            |          |
| $f_{xz^2}$       | 0.0003957  |           | $f_{xz^2}$       | -0.00025   |          | $f_{xz^2}$       | -0.00017   |          | $f_{xz^2}$       | 0.000471   |          |
| $f_{yz^2}$       | 0.0001282  |           | $f_{yz^2}$       | 0.000851   |          | $f_{yz^2}$       | -0.00036   |          | $f_{yz^2}$       | 0.000453   |          |
| $f_z$            | 0.0001651  |           | $f_z$            | -0.00023   |          | $f_z$            | -0.00033   |          | $f_z$            | -0.00018   |          |
| $f_{z(x^2-y^2)}$ | 0.0001319  |           | $f_{z(x^2-y^2)}$ | -6.5E-05   |          | $f_{z(x^2-y^2)}$ | 5.04E-05   |          | $f_{z(x^2-y^2)}$ | -0.00033   |          |
| $f_{xyz}$        | -0.000889  |           | $f_{xyz}$        | 0.000803   |          | $f_{xyz}$        | 0.000728   |          | $f_{xyz}$        | -0.00051   |          |
| $f_{(y^2-3x^2)}$ | 0.0003516  |           | $f_{(y^2-3x^2)}$ | -0.00066   |          | $f_{(y^2-3x^2)}$ | 0.001163   |          | $f_{(y^2-3x^2)}$ | -0.00137   |          |
| $f_{(x^2-3y^2)}$ | 0.0013567  |           | $f_{(x^2-3y^2)}$ | 0.001237   |          | $f_{(x^2-3y^2)}$ | 0.001186   |          | $f_{(x^2-3y^2)}$ | -0.00116   |          |
| $f_{(x^2-3y^2)}$ |            |           | $f_{(x^2-3y^2)}$ |            |          | $f_{(x^2-3y^2)}$ |            |          | $f_{(x^2-3y^2)}$ |            |          |
| $f_{xz^2}$       | -0.000359  |           | $f_{xz^2}$       | 0.000359   |          | $f_{xz^2}$       | 0.000292   |          | $f_{xz^2}$       | 0.000143   |          |
| $f_{yz^2}$       | 0.0012951  |           | $f_{yz^2}$       | 0.001213   |          | $f_{yz^2}$       | 0.001928   |          | $f_{yz^2}$       | 0.002058   |          |
| $f_z$            | 0.0009341  |           | $f_z$            | 0.001066   |          | $f_z$            | 0.00133    |          | $f_z$            | 0.001655   |          |
| $f_{z(x^2-y^2)}$ | -0.000724  |           | $f_{z(x^2-y^2)}$ | 0.000539   |          | $f_{z(x^2-y^2)}$ | 0.000348   |          | $f_{z(x^2-y^2)}$ | 0.000101   |          |
| $f_{xyz}$        | 0.0001596  |           | $f_{xyz}$        | -0.00159   |          | $f_{xyz}$        | -0.0016    |          | $f_{xyz}$        | -0.00181   |          |
| $f_{(y^2-3x^2)}$ | -0.001558  |           | $f_{(y^2-3x^2)}$ | 0.001501   |          | $f_{(y^2-3x^2)}$ | 0.000355   |          | $f_{(y^2-3x^2)}$ | 0.000237   |          |
| $f_{(x^2-3y^2)}$ | 0.0001282  |           | $f_{(x^2-3y^2)}$ | 0.000227   |          | $f_{(x^2-3y^2)}$ | 0.000413   |          | $f_{(x^2-3y^2)}$ | 0.000599   |          |

|                   |                   |           |                   |                   |          |                   |                   |          |                   |                   |          |
|-------------------|-------------------|-----------|-------------------|-------------------|----------|-------------------|-------------------|----------|-------------------|-------------------|----------|
| $f_{xyz}$         | $f_{xz^2}$        | 0.0014042 | $f_{xyz}$         | $f_{xz^2}$        | -0.00133 | $f_{xyz}$         | $f_{xz^2}$        | -0.00117 | $f_{xyz}$         | $f_{xz^2}$        | -0.00089 |
| $f_{yz^2}$        | $f_{yz^2}$        | -0.001558 |                   | $f_{yz^2}$        | 0.000753 |                   | $f_{yz^2}$        | 0.001288 |                   | $f_{yz^2}$        | 0.000869 |
| $f_z^3$           | $f_z^3$           | 0.0005932 |                   | $f_z^3$           | 0.000849 |                   | $f_z^3$           | 0.001145 |                   | $f_z^3$           | 0.001213 |
| $f_{z(x^2-y^2)}$  | $f_{z(x^2-y^2)}$  | -0.001084 |                   | $f_{z(x^2-y^2)}$  | 0.001042 |                   | $f_{z(x^2-y^2)}$  | 0.00089  |                   | $f_{z(x^2-y^2)}$  | 0.000605 |
| $f_{xyz}$         | $f_{xyz}$         | 0.0008753 |                   | $f_{xyz}$         | -0.00096 |                   | $f_{xyz}$         | -0.00095 |                   | $f_{xyz}$         | -0.00066 |
| $f_{y(y^2-3x^2)}$ | $f_{y(y^2-3x^2)}$ | -0.001211 |                   | $f_{y(y^2-3x^2)}$ | 0.001169 |                   | $f_{y(y^2-3x^2)}$ | 0.000121 |                   | $f_{y(y^2-3x^2)}$ | 0.000106 |
| $f_{x(x^2-3y^2)}$ | $f_{x(x^2-3y^2)}$ | 0.0003516 |                   | $f_{x(x^2-3y^2)}$ | 0.00047  |                   | $f_{x(x^2-3y^2)}$ | 0.00061  |                   | $f_{x(x^2-3y^2)}$ | 0.000609 |
| $f_{y(y^2-3x^2)}$ | $f_{x^2}$         | -0.000112 | $f_{y(y^2-3x^2)}$ | $f_{xz^2}$        | -0.00044 | $f_{y(y^2-3x^2)}$ | $f_{xz^2}$        | -0.00037 | $f_{y(y^2-3x^2)}$ | $f_{xz^2}$        | -0.0002  |
| $f_{yz^2}$        | $f_{yz^2}$        | 0.0001596 |                   | $f_{yz^2}$        | -0.00021 |                   | $f_{yz^2}$        | -0.00133 |                   | $f_{yz^2}$        | -0.00121 |
| $f_z^3$           | $f_z^3$           | 0.0005178 |                   | $f_z^3$           | -0.00087 |                   | $f_z^3$           | -0.00095 |                   | $f_z^3$           | -0.001   |
| $f_{z(x^2-y^2)}$  | $f_{z(x^2-y^2)}$  | -0.000464 |                   | $f_{z(x^2-y^2)}$  | -0.00026 |                   | $f_{z(x^2-y^2)}$  | -5.3E-06 |                   | $f_{z(x^2-y^2)}$  | 0.000196 |
| $f_{xyz}$         | $f_{xyz}$         | 0.0011235 |                   | $f_{xyz}$         | 0.001073 |                   | $f_{xyz}$         | 0.000934 |                   | $f_{xyz}$         | 0.000859 |
| $f_{y(y^2-3x^2)}$ | $f_{y(y^2-3x^2)}$ | 0.0008753 |                   | $f_{y(y^2-3x^2)}$ | -0.00141 |                   | $f_{y(y^2-3x^2)}$ | 0.000515 |                   | $f_{y(y^2-3x^2)}$ | 0.000519 |
| $f_{x(x^2-3y^2)}$ | $f_{x(x^2-3y^2)}$ | -0.000889 |                   | $f_{x(x^2-3y^2)}$ | 0.000642 |                   | $f_{x(x^2-3y^2)}$ | 0.000475 |                   | $f_{x(x^2-3y^2)}$ | 0.000351 |

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