Supporting information for

Modulating the magnetic properties by structural modification in a family of mixed valent Co-Ln (Ln= Gd, Dy) aggregates

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Table S1. BVS calculations for complexes 1-4.

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<td>BVS (AOS)</td>
<td>Co site</td>
<td>BVS (AOS)</td>
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<td>1.95(2)</td>
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<tr>
<td>Co2</td>
<td>2.83(3)</td>
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<td>2.83(3)</td>
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<tr>
<td>Co3</td>
<td>1.75(2)</td>
<td>Co3</td>
<td>1.75(2)</td>
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</table>

a = Assigned oxidation state

Fig. S1 TGA plots of complexes 1-4.
Fig. S2 Ball & stick model showing molecular structure of 2 in the crystal, colour code: purple, Co$^{III}$; voilet, Co$^{II}$; green, Dy; red, O; blue, N; gray, C; Hydrogen atoms are omitted for clarity.

Fig. S3 Ball & stick model showing molecular structure of 4 in the crystal, colour code: purple, Co$^{III}$; green, Dy; red, O; blue, N; gray, C; Hydrogen atoms are omitted for clarity.
**Fig. S4** Field-dependencies of isothermal normalized magnetizations for complex 1 collected for temperatures ranging from 2-10 K.

**Fig. S5** Field-dependencies of isothermal normalized magnetizations for complex 3 collected for temperatures ranging from 2-10 K.

**Fig. S6** $M/N\mu_B$ vs $H/T$ plots for complex 1 at 2-10 K.
**Fig. S7** M/N$_{\text{ÎB}}$ vs H/T plots for complex 3 at 2-10 K

**Fig. S8** Field-dependencies of isothermal normalized magnetizations for complex 2 collected for temperatures ranging from 2-10 K

**Fig. S9** Field-dependencies of isothermal normalized magnetizations for complex 4 collected for temperatures ranging from 2-10 K.
Fig. S10 M/NμB vs H/T plots for complex 2 at 2-10 K.

Fig. S11 M/NμB vs H/T plots for complex 4 at 2-10 K.

Fig. S12 Frequency dependence of the in-phase (χ') ac susceptibility for complex 2 under zero dc field
Fig. S13 Frequency dependence of the out of phase ($\chi''$) ac susceptibility for complex 2 under zero dc field.

Fig. S14 Temperature dependence of the in-phase ($\chi'$) ac susceptibility for complex 4 under zero dc field.

Fig. S15 Temperature dependence of the out of phase ($\chi''$) ac susceptibility for complex 4 under zero dc field.
Fig. S16 Frequency dependence of the out of phase ($\chi''$) ac susceptibility for complex 4 under zero dc field.