Electronic Supporting Information

# The New Dicyanoruthenium(III) Building Block with 2'-Hydroxyacetophenone Imine for Heterobimetallic Complexes

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**Caption of Content**

1. **Figure S1.** The 2D layer of **4** constructed from the interchain $\pi$-$\pi$ interactions (rose dashes) between the aromatic rings of the nappa ligand.

2. **Figure S2**. $1/\chi_M$(■) for complex **1** at 1 kOe.

3. **Figure S3.** Field dependence of the magnetization for **1**, the lines represent the Brillouin function that correspond to $S = 1/2+2$ with $g = 2.0$.

4. **Figure S4.** Field dependence of the magnetization for **2**, the lines represent the Brillouin function that correspond to $S = 1/2+2$ with $g = 2.0$.

5. **Figure S5.** Temperature dependence of the in-phase $\chi'$ (top) and out-of-phase $\chi''$ (bottom) at different frequencies in 2 Oe ac field oscillating at 666 Hz with zero applied dc field for **3**.

6. **Figure S6.** $1/\chi_M$ vs $T$ (■) for complex **4** at 1 kOe.

7. **Figure S7.** Thermal dependence of the magnetic susceptibility at $T \leq 8$ K for complex **4** at 1 kOe.

8. **Figure S8.** $1/\chi_M$ vs T (■) for complex **5** at 1 kOe.

9. **Figure S9**. Field dependence of the magnetization for **5**, the lines represent the Brillouin function that correspond to $S = 1/2+2$ with $g = 2.0$.

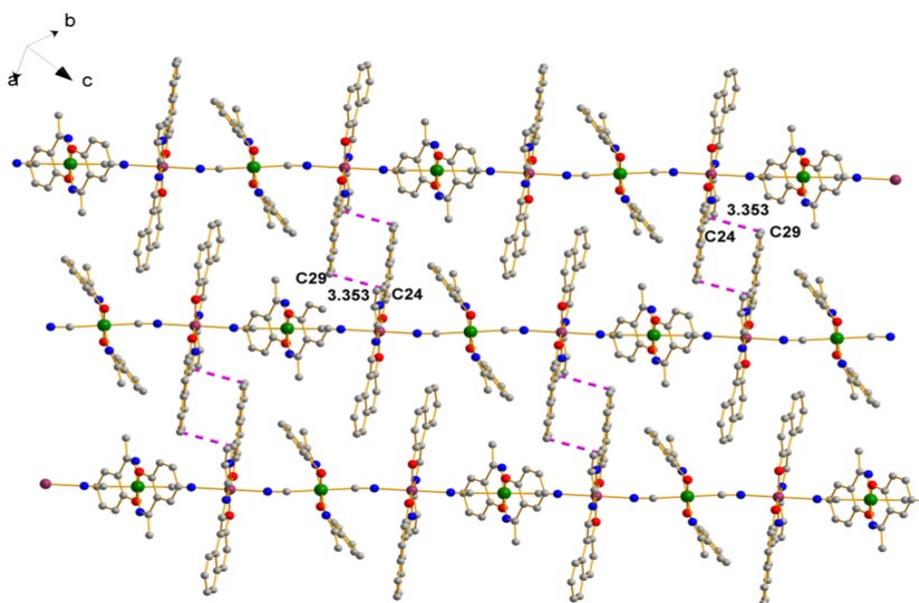10. **Figure S10**. IR spectra of complexes **1**- **5**.

**Figure S1**. The 2D layer of **4** constructed from the interchain π-π interactions (rose dashes) between the aromatic rings of the nappa ligand.
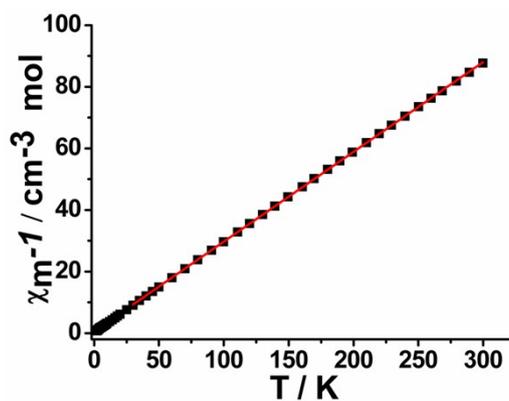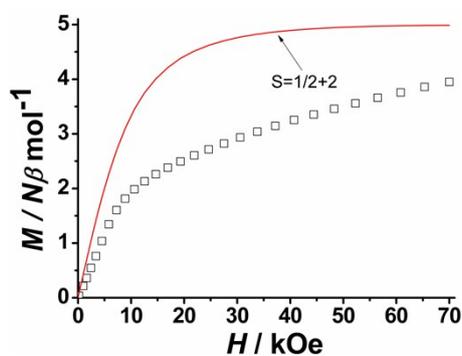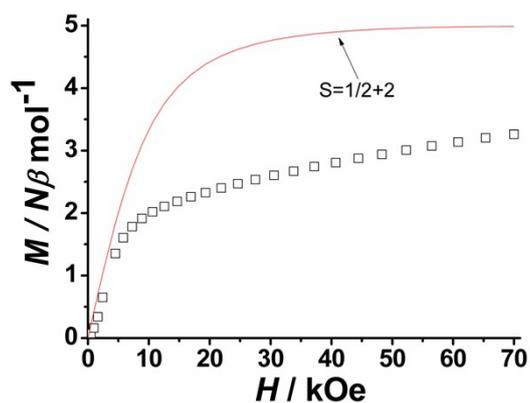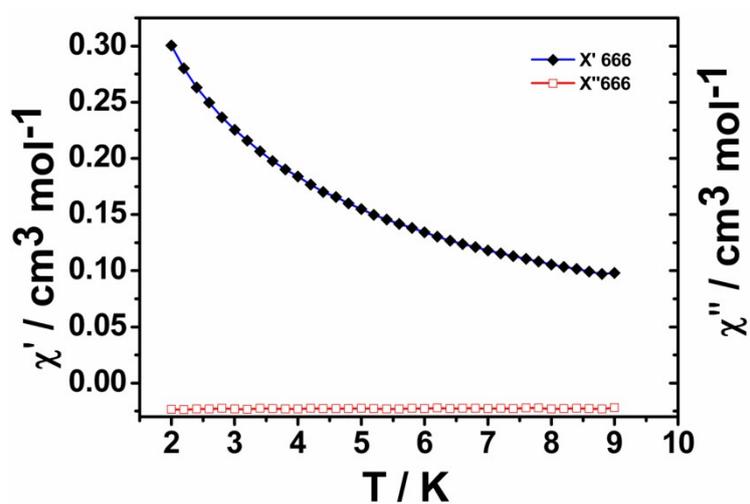
**Figure S4.** Field dependence of the magnetization for **2**, the lines represent the Brillouin function that correspond to $S = 1/2+2$ with $g = 2.0$.



**Figure S5**. Temperature dependence of the in-phase $\chi'$ (top) and out-of-phase $\chi''$ (bottom) at different frequencies in 2 Oe ac field oscillating at 666 Hz with zero applied dc field for **3**.
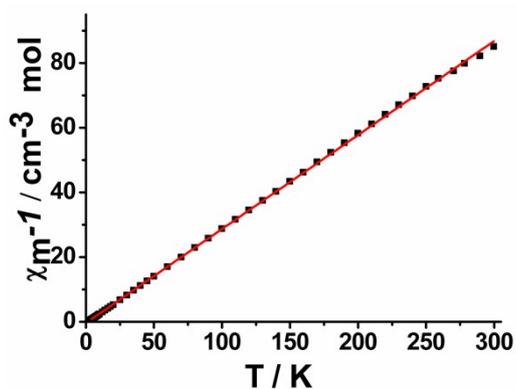


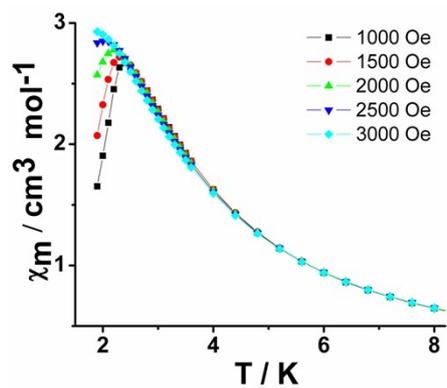**Figure S6.** $1/\chi_M$ (■) vs $T$ for complex **4** at 1 kOe

**Figure S7.** Thermal dependence of the magnetic susceptibility at $T \leqslant 8$ K for complex **4** at 1 kOe.
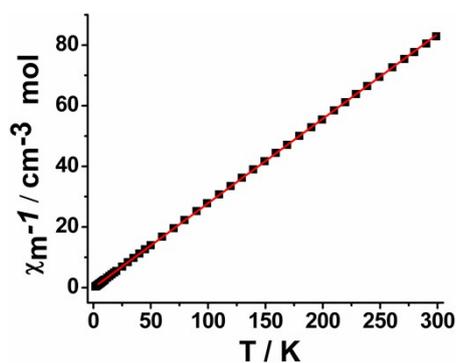


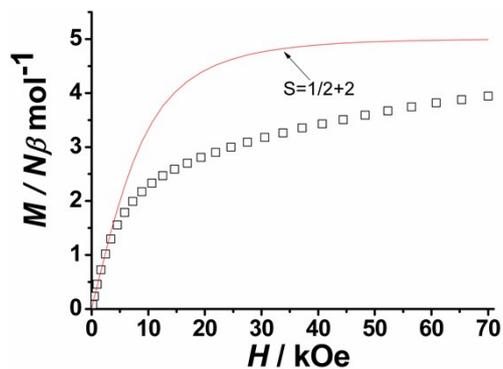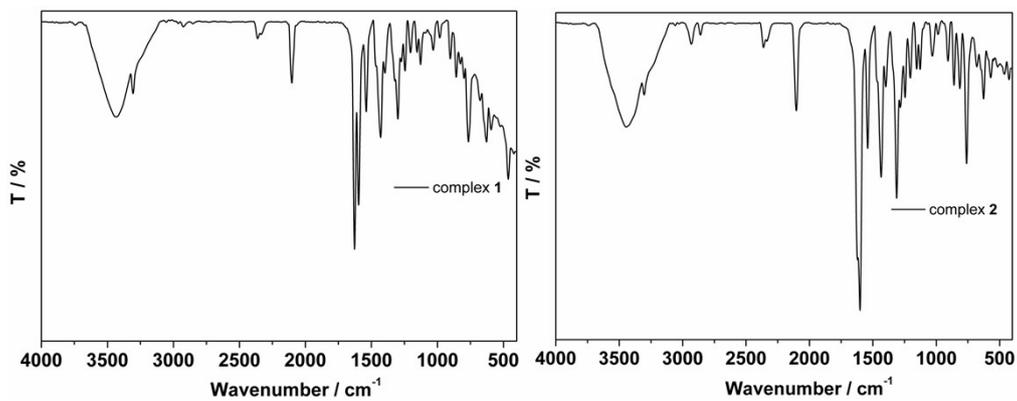**Figure S8.** $1/\chi_M$(■) vs $T$ for complex **5** at 1 kOe.



**Figure S9.** Field dependence of the magnetization for **5**, the lines represent the Brillouin function that correspond to $S = 1/2+2$ with $g = 2.0$.
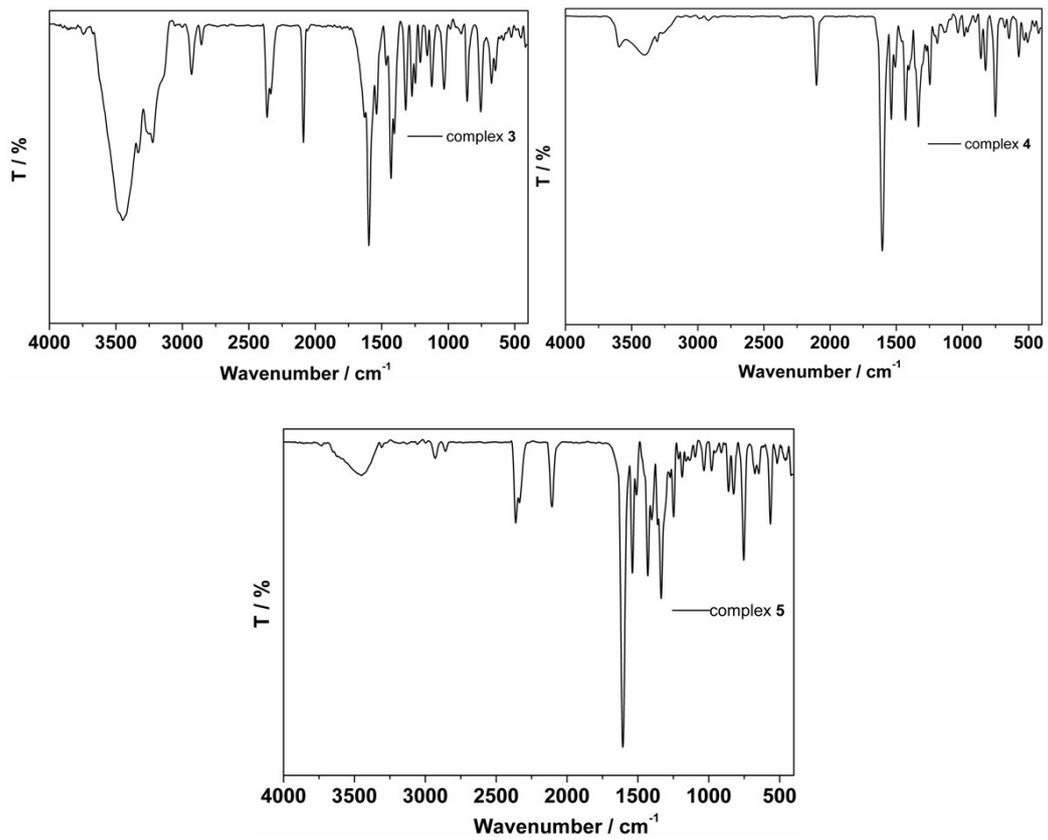
**Figure S10.** IR spectra for complexes **1-5**.