

## Supporting Information

**Charge-transfer two-dimensional layers constructed from a 2:1 assembly of paddlewheel diruthenium(II, II) complexes and bis[1,2,5]dithiazolotetracyanoquinodimethane: Bulk magnetic behavior as a function of inter-layer interactions**

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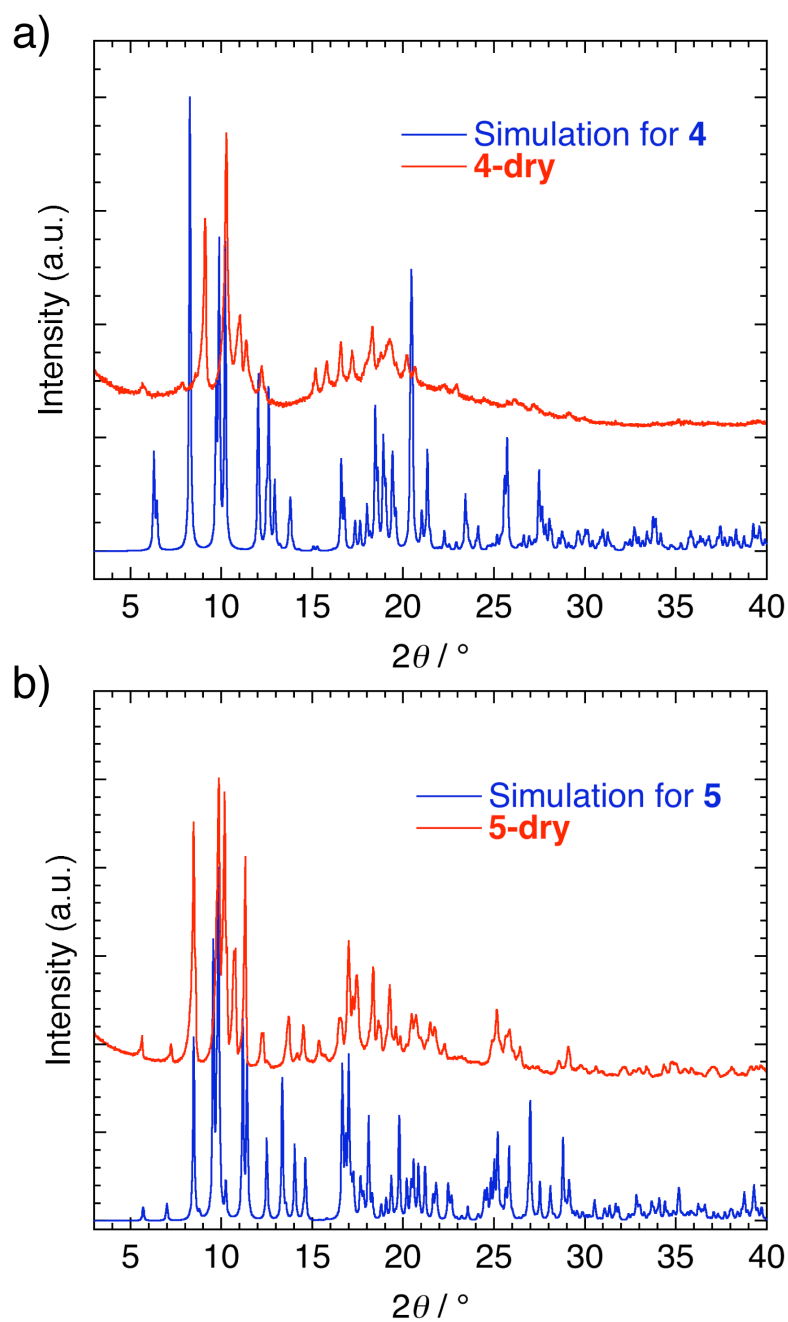


Fig. S1. XRPD patterns of **4-dry** (a) and **5-dry** (b) with simulated patterns of fresh samples **4** and **5**, where 4-dry and 5-dry were prepared by heating at 150°C.

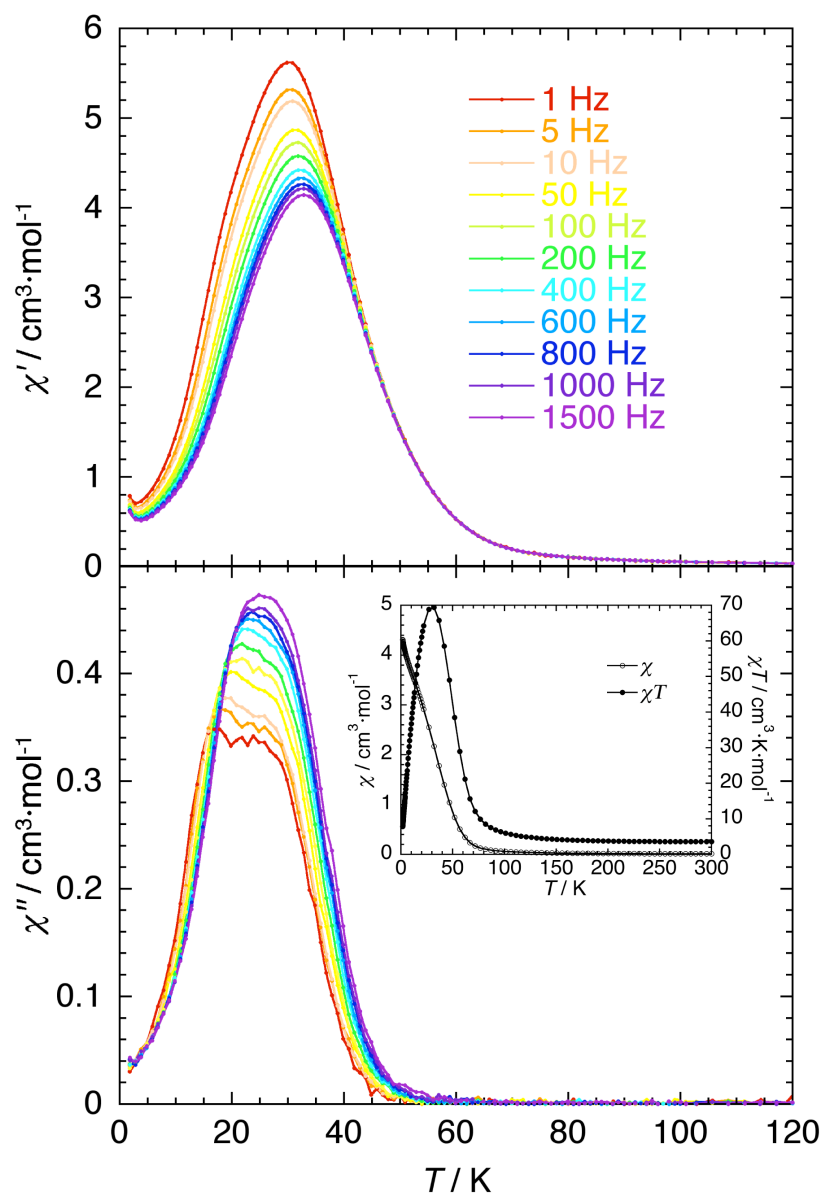


Fig. S2. Temperature dependence of ac susceptibilities ( $\chi'$ : in-phase and  $\chi''$ : out-of-phase) of dried sample of **4** (**4-dry**) prepared by removing the crystallization solvents heating up to 150°C (zero dc field and 3 Oe oscillation field). Inset: Temperature dependence of dc susceptibility ( $\chi$ ) and  $\chi T$  of **4-dry** measured at 1 kOe applied field.

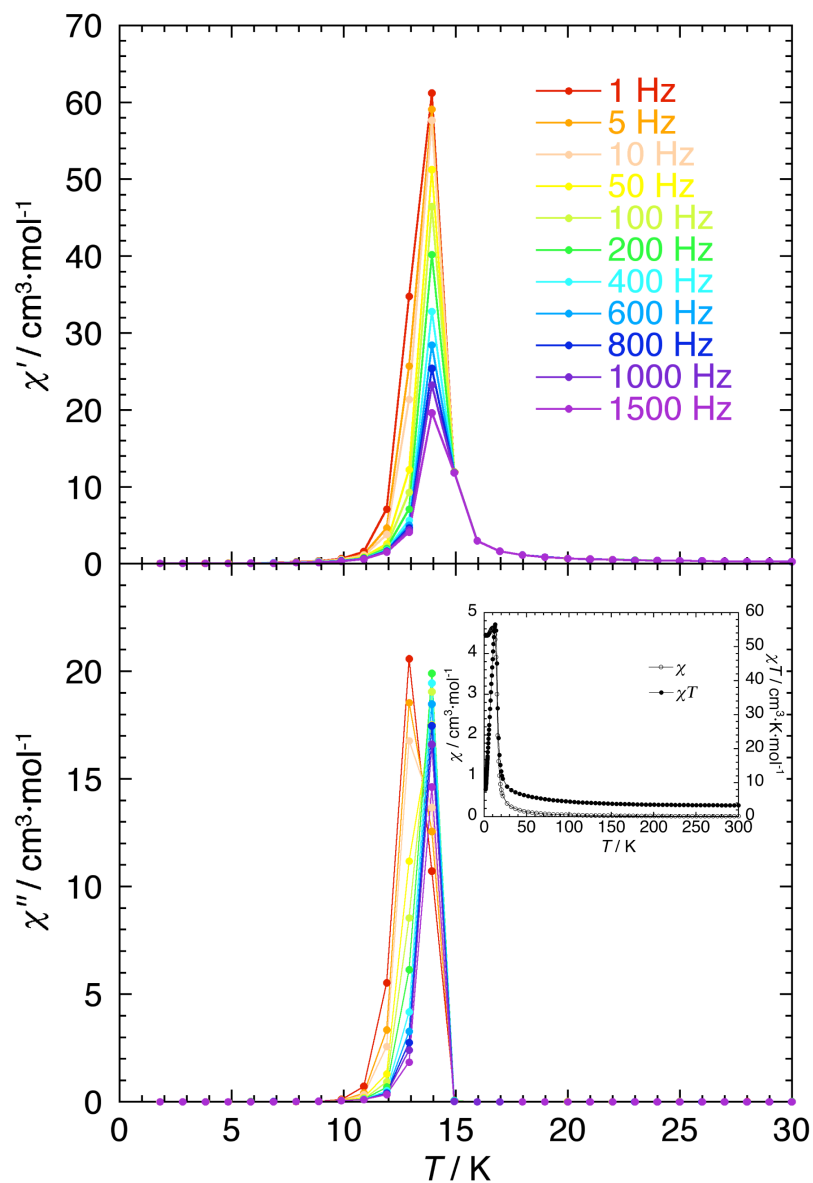


Fig. S3. Temperature dependence of ac susceptibilities ( $\chi'$ : in-phase and  $\chi''$ : out-of-phase) of dried sample of **5** (**5-dry**) prepared by removing the crystallization solvents heating up to 150°C (zero dc field and 3 Oe oscillation field). Inset: Temperature dependence of dc susceptibility ( $\chi$ ) and  $\chi T$  of **5-dry** measured at 1 kOe applied field.