

Electronic Supplementary Information

Study on the Relationship between Magnetic Field and Dielectric Properties in Two Ferromagnetic complexes

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Table S1. Metal–Ligand Bond Lengths (Å) and Angles (°) in Complex 1.

| | | | | | |
|-----------------------------------------|------------|------------------------------------------|------------|------------------------------------------|------------|
| Cr1–O1 ⁱ | 1.9796(11) | Cr1–O1 ^{iv} | 1.9796(11) | Fe1–O2 ^{vii} | 2.1174(13) |
| Cr1–O1 ⁱⁱ | 1.9796(11) | Cr1–O1 ^v | 1.9796(11) | Fe1–O2 ^{viii} | 2.1175(13) |
| Cr1–O1 ⁱⁱⁱ | 1.9796(11) | Fe1–O2 | 2.1174(13) | Fe1–O2 ^{ix} | 2.1175(13) |
| Cr1–O1 | 1.9796(11) | Fe1–O2 ^{vi} | 2.1174(13) | Fe1–O2 ^x | 2.1175(13) |
| O1 ⁱ –Cr1–O1 ⁱⁱ | 180.00 | O1 ⁱ –Cr1–O1 ^v | 88.92(5) | O2 ^{vii} –Fe1–O2 ^{ix} | 88.76(8) |
| O1 ⁱ –Cr1–O1 ⁱⁱⁱ | 91.08(5) | O1 ⁱⁱ –Cr1–O1 ^v | 91.08(5) | O2–Fe1–O2 ^x | 93.10(6) |
| O1 ⁱⁱ –Cr1–O1 ⁱⁱⁱ | 88.92(5) | O1 ⁱⁱ –Cr1–O1 ^v | 180.0 | O2 ^{vi} –Fe1–O2 ^{ix} | 93.10(6) |
| O1 ⁱ –Cr1–O1 | 88.92(5) | O1 ^{iv} –Cr1–O1 | 180.0 | O2 ^{vii} –Fe1–O2 ^{ix} | 88.76(8) |
| O1 ⁱⁱ –Cr1–O1 | 91.08(5) | O2–Fe1–O2 ^{viii} | 85.09(7) | O2 ^{viii} –Fe1–O2 ^{ix} | 93.10(6) |
| O1 ⁱⁱⁱ –Cr1–O1 | 88.92(5) | O2–Fe1–O2 ^{ix} | 177.48(7) | O2–Fe1–O2 ^x | 93.10(6) |
| O1 ⁱ –Cr1–O1 ^{iv} | 91.08(5) | O2 ^{vi} –Fe1–O2 ^{ix} | 93.11(6) | O2 ^{vi} –Fe1–O2 ^x | 177.47(7) |
| O1 ⁱⁱ –Cr1–O1 ^{iv} | 88.92(5) | O2–Fe1–O2 ^{viii} | 85.09(7) | O2 ^{vii} –Fe1–O2 ^{ix} | 88.76(8) |
| O1 ⁱⁱⁱ –Cr1–O1 ^{iv} | 91.08(5) | O2 ^{vi} –Fe1–O2 ^{viii} | 93.10(6) | O2 ^{viii} –Fe1–O2 ^x | 88.76(8) |

Symmetry codes: (i) $y-1, -x+y, -z$; (ii) $-y+1, x-y+2, z$; (iii) $x-y+1, x+1, -z$; (iv) $-x, -y+2, -z$; (v) $-x+y-1, -x+1, z$; (vi) $-y+1, -x+1, -z+1/2$; (vii) $-x+y, -x+1, z$; (viii) $x, x-y+1, -z+1/2$; (ix) $-x+y, y, -z+1/2$; (x) $-y+1, x-y+1, z$; (xi) $-y+2, x-y+1, z$; (xii) $-x+y+1, -x+1, z$.

Table S2. Metal–Ligand Bond Lengths (Å) and Angles (°) in Complex 2.

| | | | | | |
|------------------------------------------|------------|-------------------------------------------|------------|------------------------------------------|------------|
| Cr1–O1 | 1.9753(15) | Cr1–O1 ^{xi} | 1.9753(15) | Ni1–O2 ^{iv} | 2.0530(16) |
| Cr1–O1 ^{viii} | 1.9753(15) | Cr1–O1 ^{xii} | 1.9753(15) | Ni1–O2 ^v | 2.0530(16) |
| Cr1–O1 ^{ix} | 1.9753(15) | Ni1–O2 | 2.0530(16) | Ni1–O2 ^{vi} | 2.0530(16) |
| Cr1–O1 ^x | 1.9753(15) | Ni1–O2 ⁱⁱⁱ | 2.0530(16) | Ni1–O2 ^{vii} | 2.0530(16) |
| O1 ^{viii} –Cr1–O1 ^x | 91.39(7) | O1 ^{viii} –Cr1–O1 ^{xii} | 91.39(7) | O2 ⁱⁱⁱ –Ni1–O2 | 176.63(10) |
| O1 ^{viii} –Cr1–O1 ^{ix} | 180.00(8) | O1 ^{ix} –Cr1–O1 ^{xii} | 88.61(7) | O2 ^{iv} –Ni1–O2 ^{vi} | 93.20(7) |
| O1 ^{ix} –Cr1–O1 ^x | 88.61(7) | O1 ^x –Cr1–O1 ^{xii} | 91.39(7) | O2 ^v –Ni1–O2 ^{vi} | 176.63(10) |
| O1 ^{viii} –Cr1–O1 ^{xi} | 88.61(7) | O1 ^{xi} –Cr1–O1 ^{xii} | 180.00(8) | O2 ⁱⁱⁱ –Ni1–O2 ^{vi} | 89.31(10) |
| O1 ^{ix} –Cr1–O1 ^{xi} | 91.39(7) | O1–Cr1–O1 ^{xii} | 88.61(7) | O2–Ni1–O2 ^{vi} | 93.19(7) |
| O1 ^x –Cr1–O1 ^{xi} | 88.61(7) | O2 ^{iv} –Ni1–O2 ^v | 89.31(10) | O2 ^{iv} –Ni1–O2 ^{vii} | 176.63(10) |
| O1 ^{viii} –Cr1–O1 | 88.61(7) | O2 ⁱⁱⁱ –Ni1–O2 ⁱⁱⁱ | 84.40(10) | O2 ^v –Ni1–O2 ^{vii} | 93.19(7) |
| O1 ^{ix} –Cr1–O1 | 91.39(7) | O2 ^v –Ni1–O2 ⁱⁱⁱ | 93.20(7) | O2 ⁱⁱⁱ –Ni1–O2 ^{vii} | 93.19(7) |
| O1 ^x –Cr1–O1 | 180.00(8) | O2 ^{iv} –Ni1–O2 | 93.20(7) | O2–Ni1–O2 ^{vii} | 89.31(10) |
| O1 ^{xi} –Cr1–O1 | 91.39(7) | O2 ^v –Ni1–O2 | 84.40(10) | O2 ^{vi} –Ni1–O2 ^{vii} | 84.40(10) |

Symmetry codes: (i) $-y+1, x-y, z$; (ii) $-x+y+1, -x+1, z$; (iii) $-y+1, -x+1, -z+1/2$; (iv) $-y+1, x-y+1, z$; (v) $-x+y, y, -z+1/2$; (vi) $-x+y, -x+1, z$; (vii) $x, x-y+1, -z+1/2$; (viii) $x-y+1, x, -z$; (ix) $-x+y+1, -x+2, z$; (x) $-x+2, -y+2, -z$; (xi) $-y+2, x-y+1, z$; (xii) $y, -x+y+1, -z$.

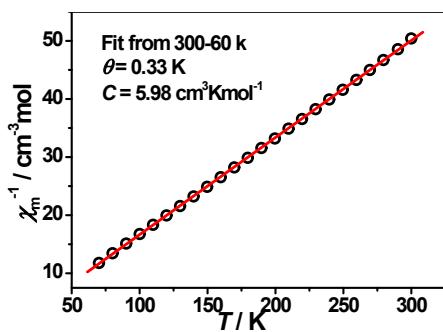


Fig. S1 Plot of χ_M^{-1} vs T for **1**. The solid line represents the best-fit curve following Curie-Weiss law.

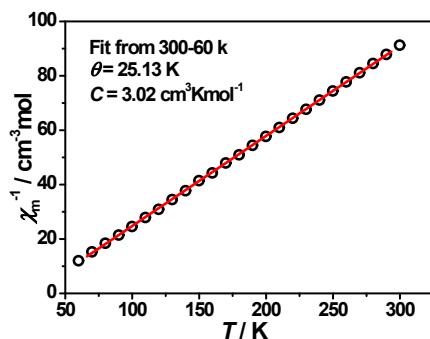


Fig. S2 Plot of χ_M^{-1} vs T for **2**. The solid line represents the best-fit curve following Curie-Weiss law.

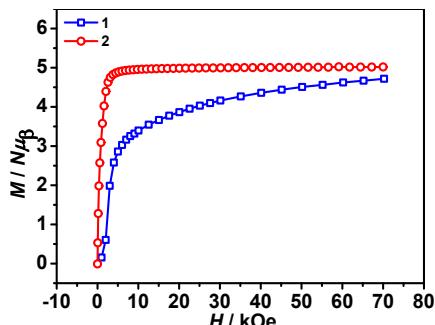


Fig. S3 Plot of M vs H for **1** and **2** at 2 K.

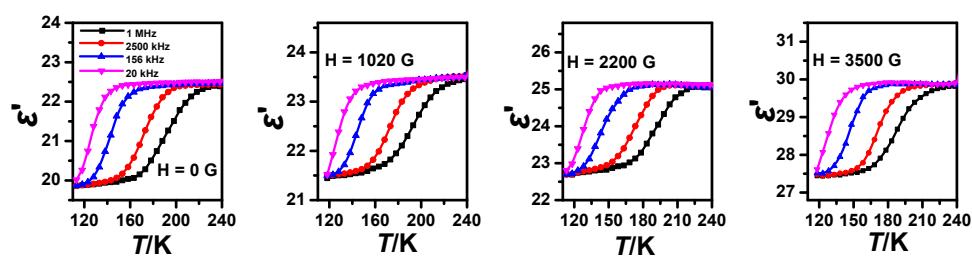


Fig. S4 Temperature dependence of the real ϵ' part of the dielectric permittivity in the case of no field and different external field for **1**.

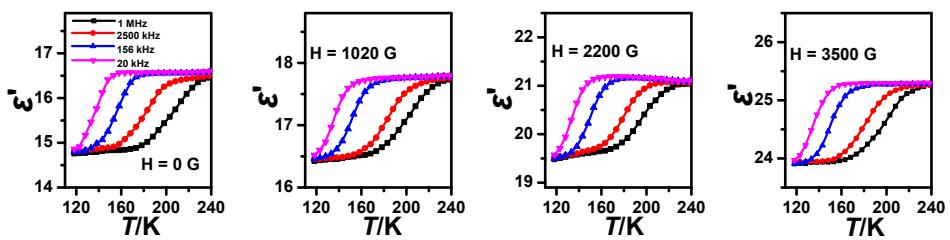


Fig. S5 Temperature dependence of the real ϵ' part of the dielectric permittivity in the case of no field and different external field for **2**.

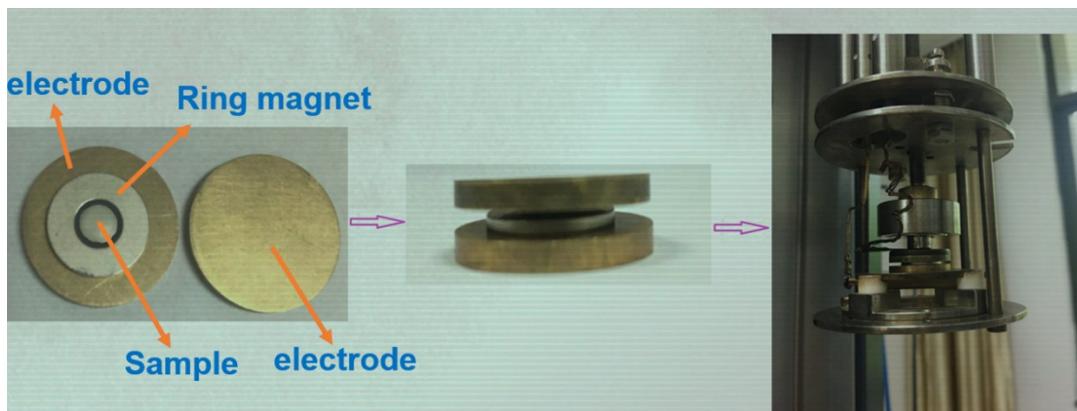


Fig. S6 The model of dielectric permittivity measurements for **1** and **2** under an applied magnetic field.

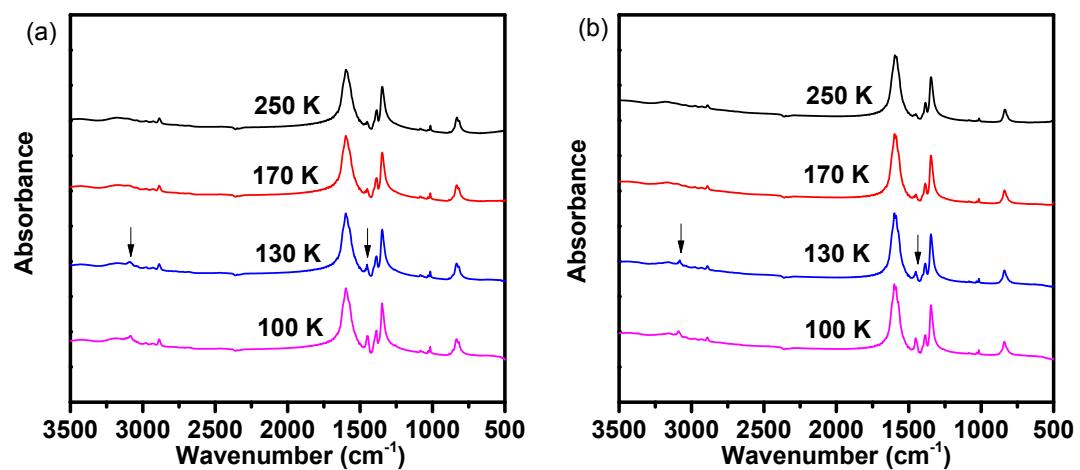


Fig. S7 Details of the IR spectra corresponding to spectral range 500-3500 cm^{-1} for (a) **1** and (b) **2**.

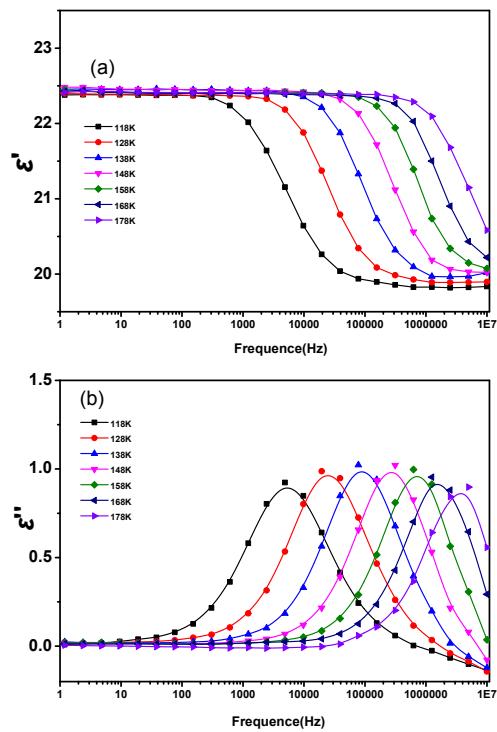


Fig. S8 Frequency dependence of (a) the real ϵ' part and (b) the imaginary ϵ'' of the dielectric permittivity for **1**.

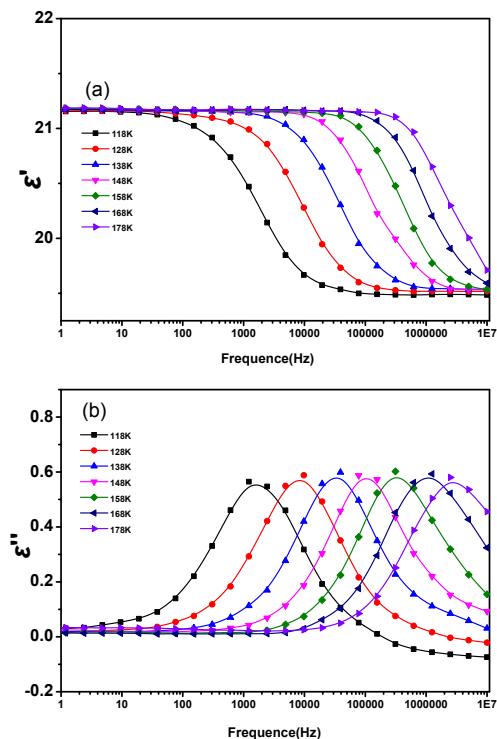


Fig. S9 Frequency dependence of (a) the real ϵ' part and (b) the imaginary ϵ'' of the dielectric permittivity for **2**.

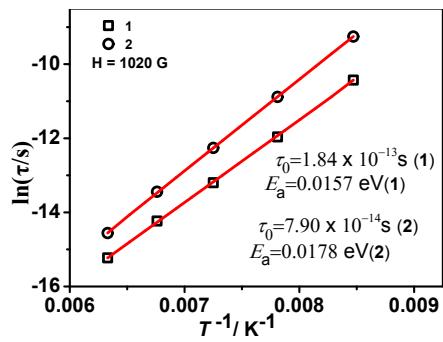


Fig. S10 Arrhenius plots of $\ln(\tau)$ vs the inverse temperature T^{-1} under an applied magnetic field of 1020 G for **1** and **2**. Red lines show the fit of data to the Arrhenius expression $\tau = \tau_0 \exp(E_a / k_B T)$.

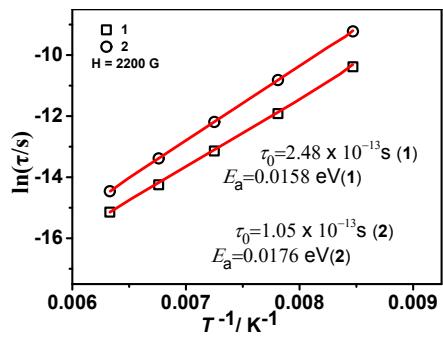


Fig. S11 Arrhenius plots of $\ln(\tau)$ vs the inverse temperature T^{-1} under an applied magnetic field of 2200 G for **1** and **2**. Red lines show the fit of data to the Arrhenius expression $\tau = \tau_0 \exp(E_a / k_B T)$.

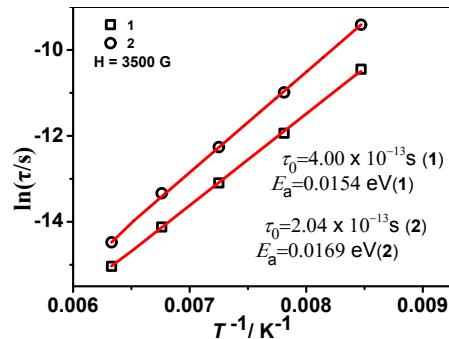


Fig. S12 Arrhenius plots of $\ln(\tau)$ vs the inverse temperature T^{-1} under an applied magnetic field of 3500 G for **1** and **2**. Red lines show the fit of data to the Arrhenius expression $\tau = \tau_0 \exp(E_a / k_B T)$.

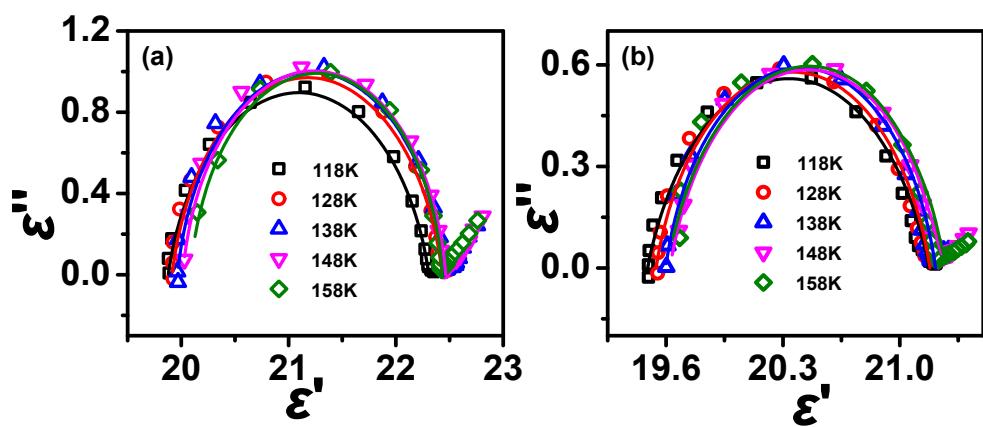


Fig. S13 Cole-Cole diagram of **1** (a) and **2** (b), plotted using χ_M' and χ_M'' at different temperature. The solid lines represent the fits to a general Debye model.