

SUPPORTING INFORMATION

Investigation of the Slow Relaxation of the Magnetization Dynamics in Homoleptic Ene-Diamido Organodysprosium(III) Complexes with K⁺/Arene Interactions.

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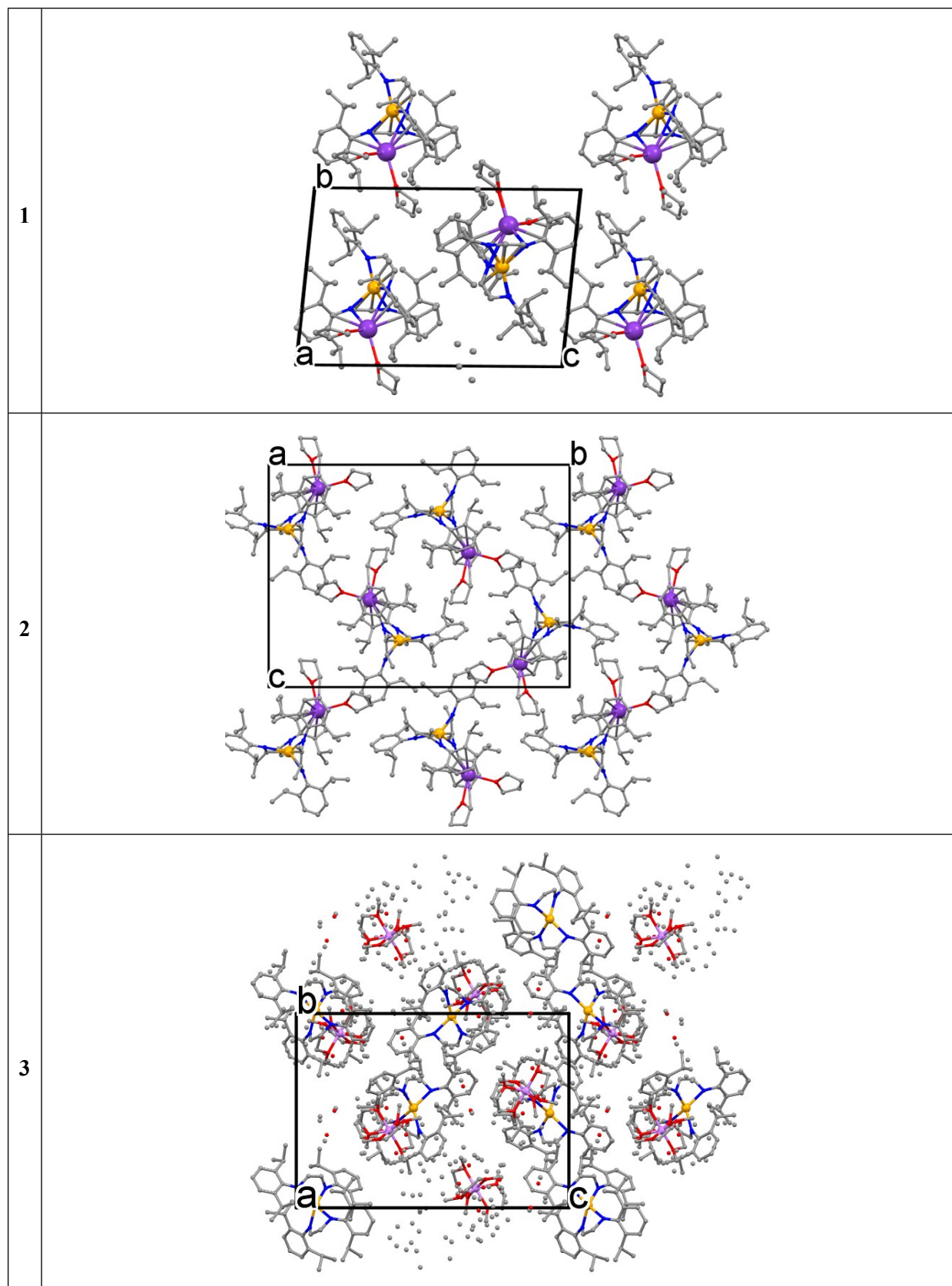


Figure S1: Perspective view of the crystal packing for compounds **1**, **2**, and **3** along the *a* crystallographic axis. Hydrogen atoms have been omitted for clarity.

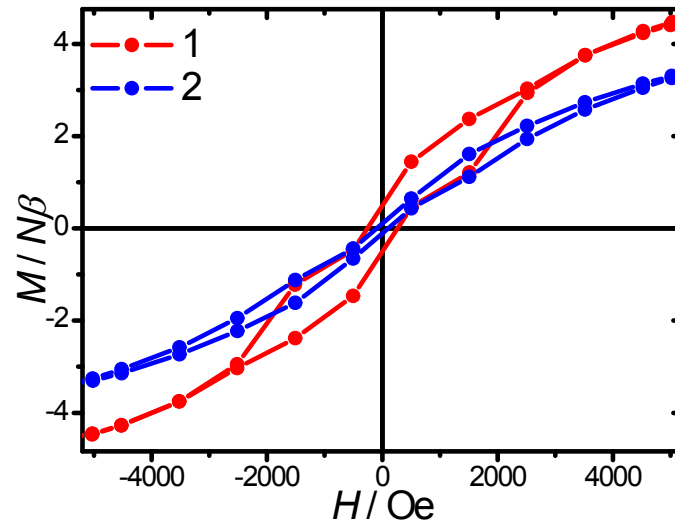


Figure S2: Hysteresis loops obtained at 1.8 K for 1 and 2 at an average sweep rate of 22 Oe.s⁻¹.

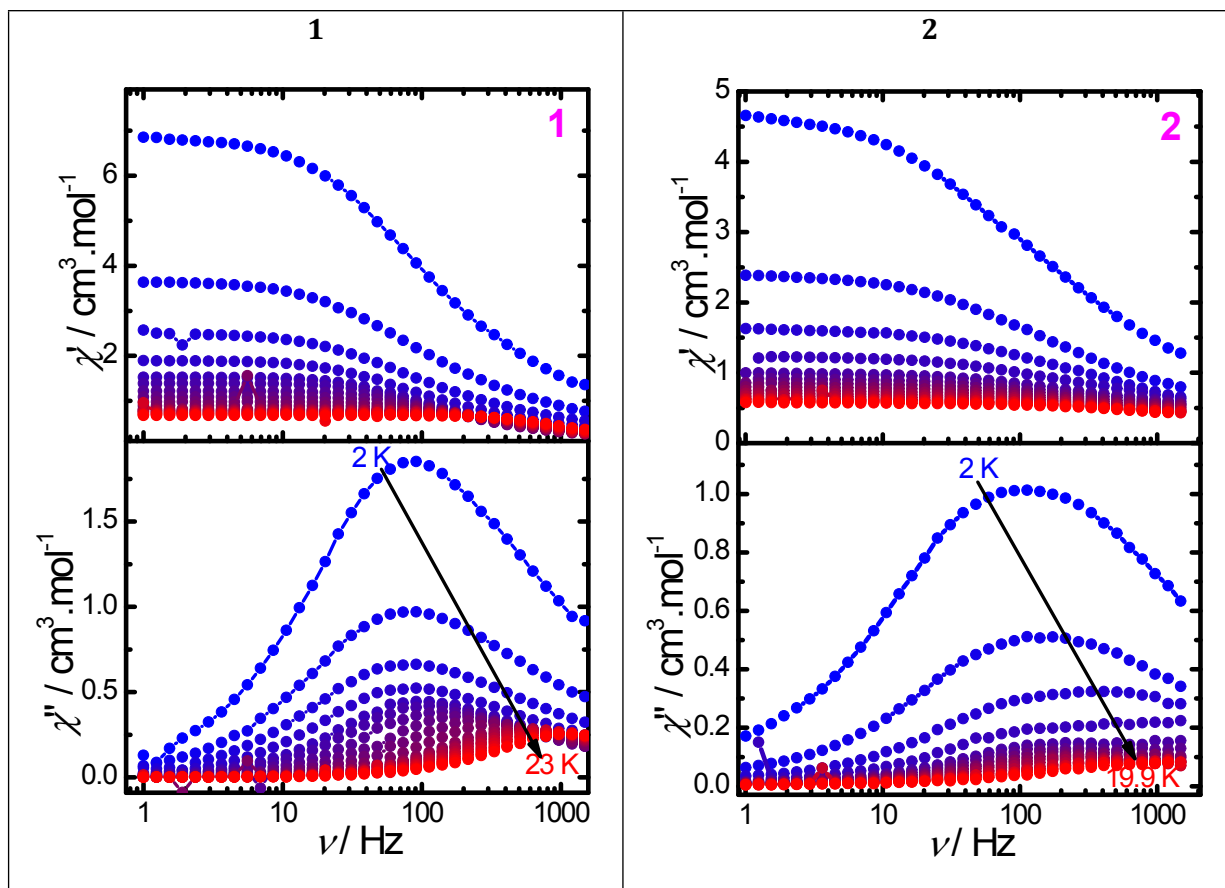


Figure S3: Frequency dependence of χ' and χ'' for 1 and 2 under a zero-dc field.

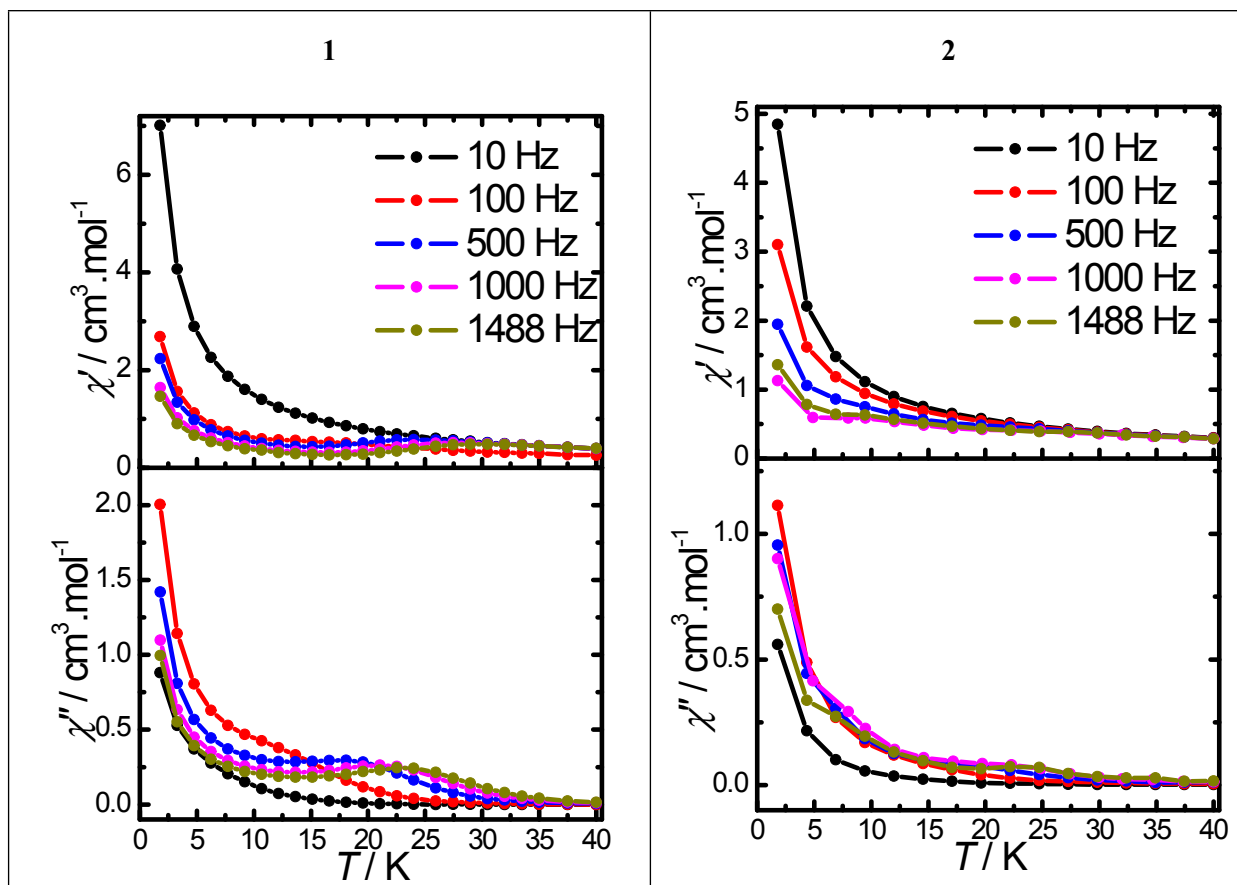


Figure S4: Temperature dependence of χ' and χ'' for **1** and **2** for different frequencies under a zero-dc field.

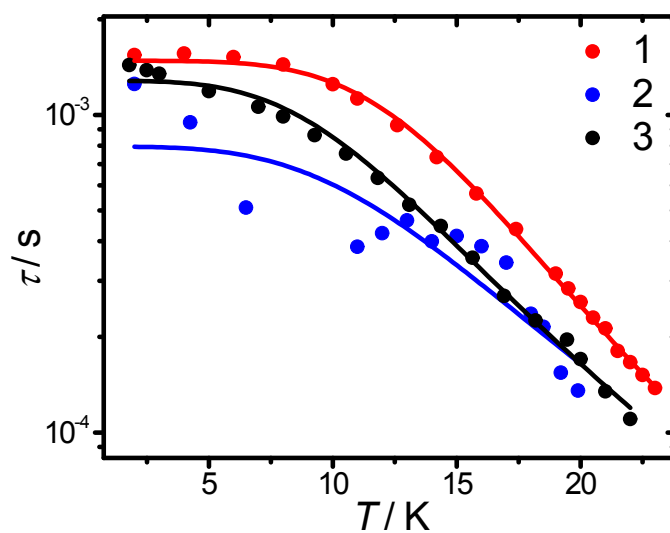


Figure S5: Temperature dependence of the relaxation time, τ , for **1-3** under a zero dc-field. The solid magenta lines correspond to the fit with Eq. 2.

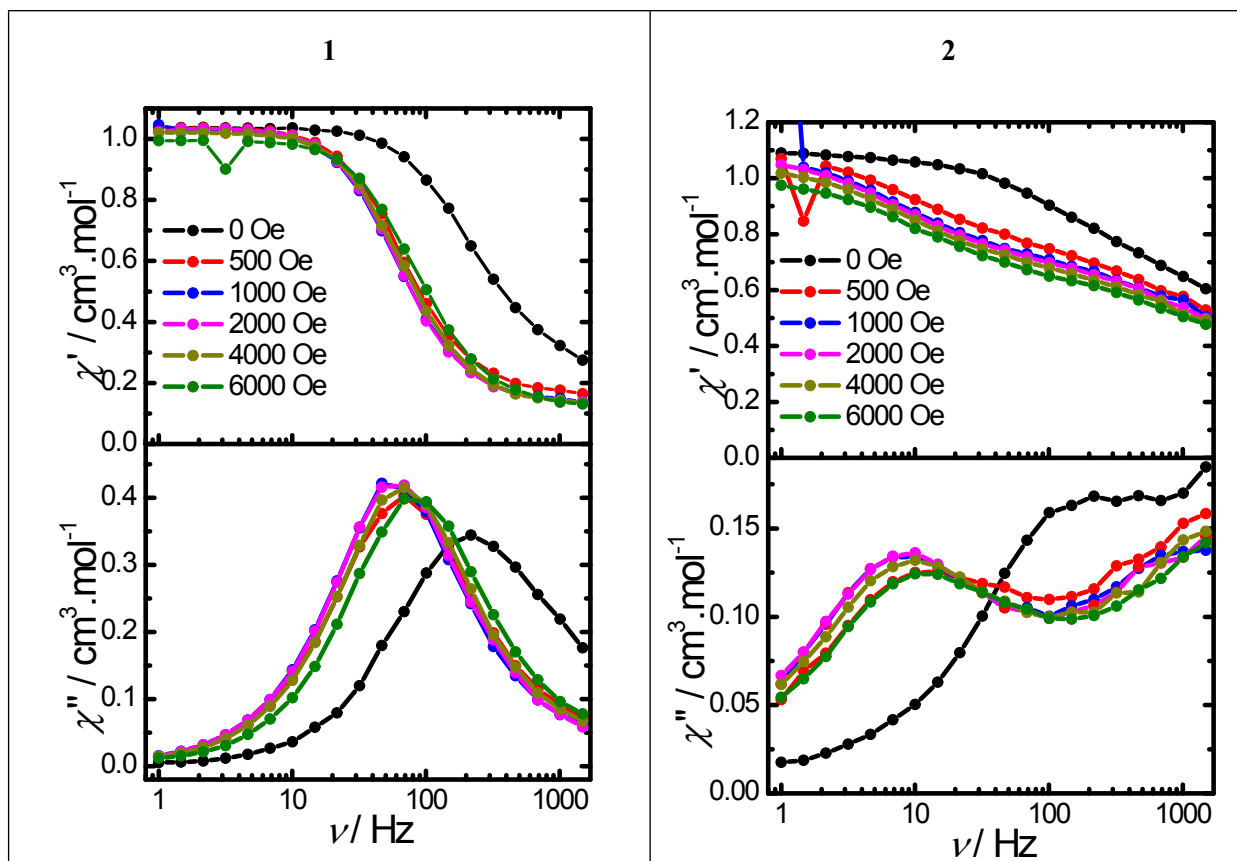


Figure S6: Frequency dependence of χ' and χ'' for **1** (15 K) and **2** (10 K) for various dc-fields.

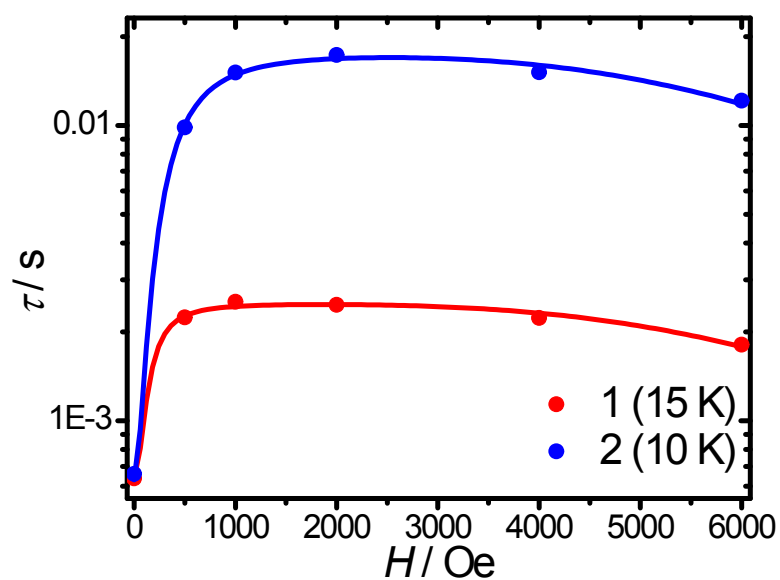


Figure S7: Field dependence of the relaxation time for **1** (15 K) and **2** (10 K). The solid lines represent the fit with Equation 3.

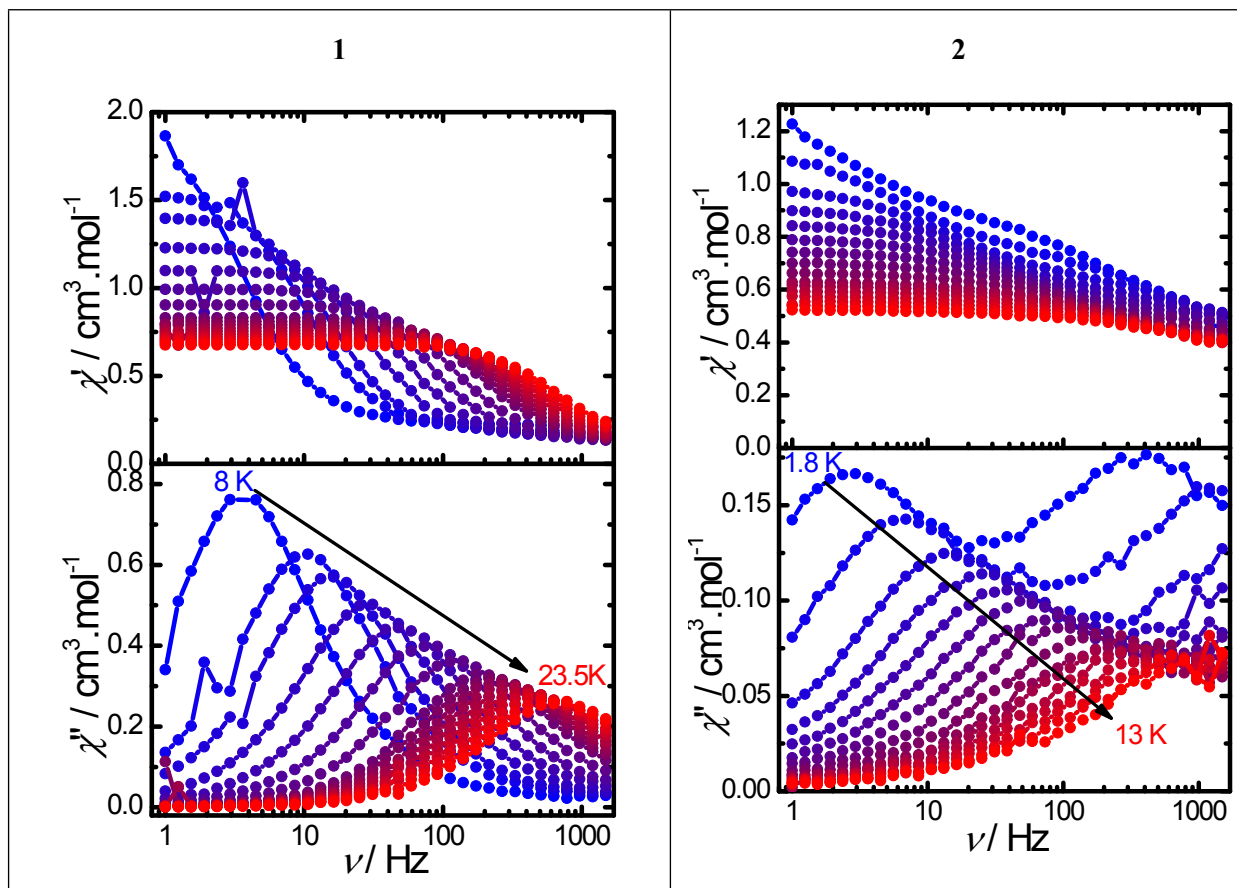


Figure S8: Frequency dependence of χ' and χ'' for **1** (1000 Oe) and **2** (2000 Oe).

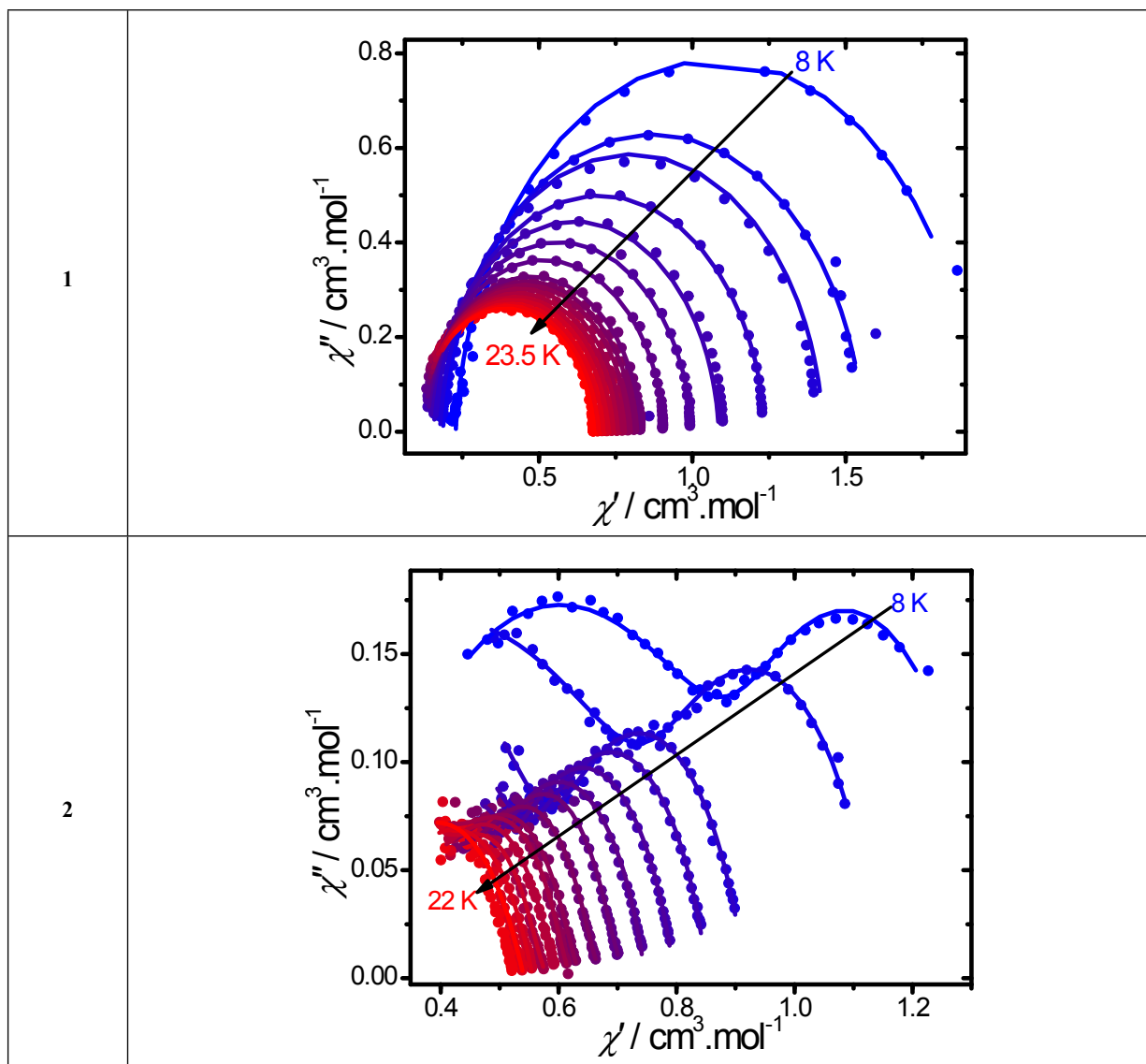


Figure S9: Cole-Cole (Argand) plot obtained using the ac susceptibility data for **1** and **2**. The solid lines correspond to the best fit obtained with a generalized Debye model (**1**) or with the sum of two modified Debye functions (**2**).

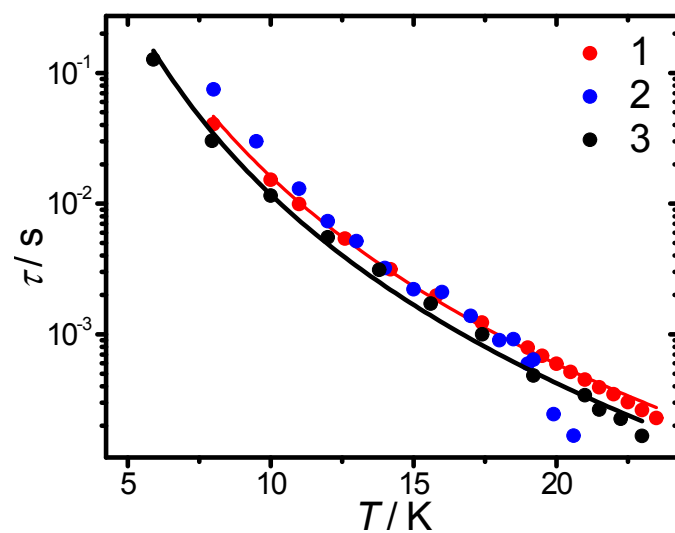


Figure S10: Temperature dependence of the relaxation time for 1-3 using the ac susceptibility data in the presence of a dc field. The solid lines represent the fit with a Raman process.

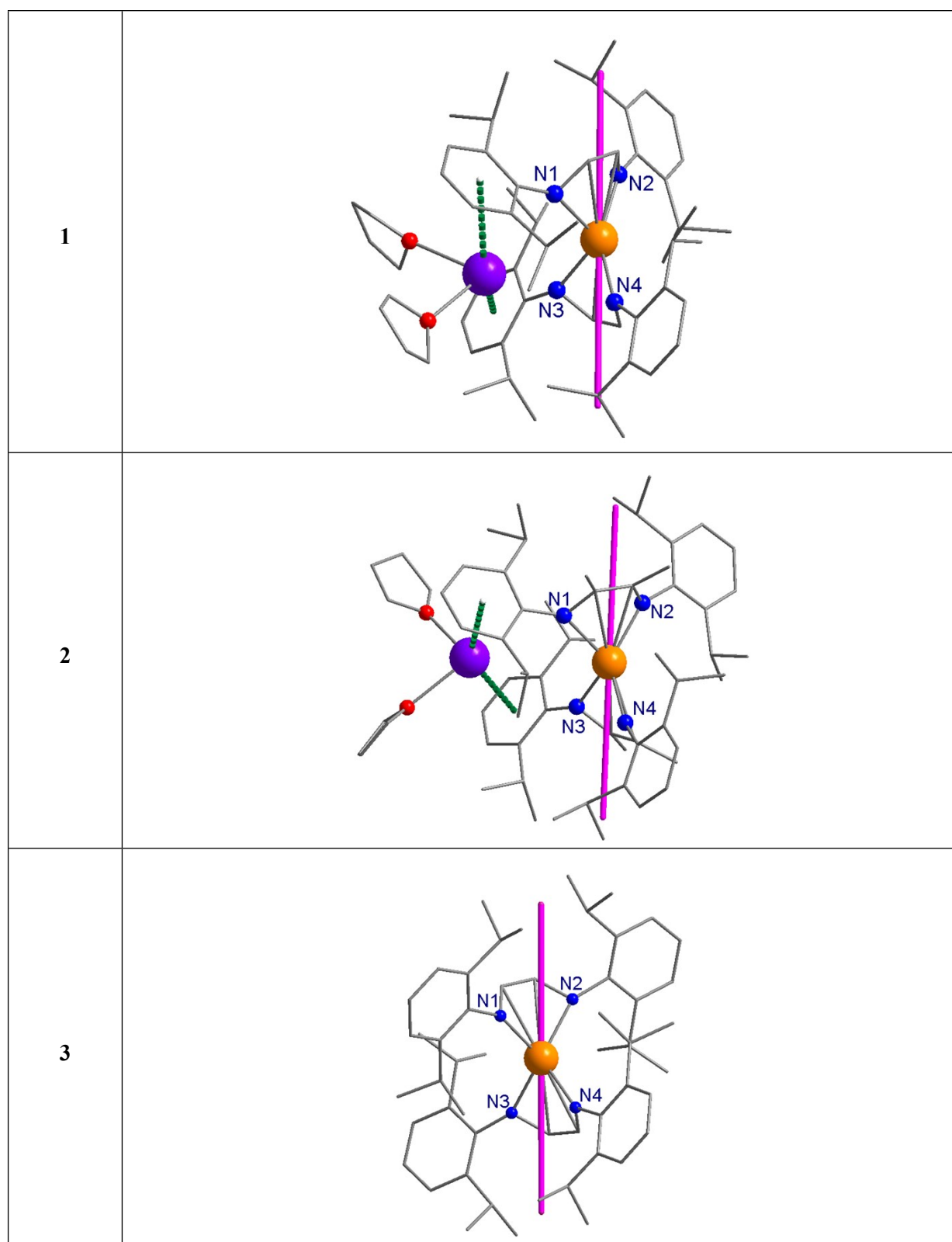


Figure S11: Orientation of the anisotropic axis (purple) in **1** obtained from the MAGELLAN¹ software.

Table S1: Crystallographic data and structure refinement details for 1 and 2.

| Compound | 1 | 2 |
|--|---|---|
| Empirical formula | C ₆₀ H ₈₈ DyKN ₄ O ₂ | C ₆₄ H ₉₆ DyKN ₄ O ₂ |
| Formula weight | 1098.94 | 1155.04 |
| <i>T</i> , K | 120 | 200 |
| Crystal system | Triclinic | Monoclinic |
| Space group | <i>P</i> -1 | <i>P</i> 2 ₁ / <i>n</i> |
| <i>a</i> , Å | 11.7767(7) | 10.98330(10) |
| <i>b</i> , Å | 13.3468(11) | 27.4951(3) |
| <i>c</i> , Å | 19.3209(11) | 20.5502(2) |
| α , deg | 81.5632(16) | 90 |
| β , deg | 79.7047(11) | 98.2020(10) |
| γ , deg | 72.2832(9) | 90 |
| <i>V</i> , Å ³ | 2832.4(3) | 6142.41(11) |
| <i>Z</i> | 2 | 4 |
| <i>d</i> _{calcd} , g/cm ³ | 1.289 | 1.249 |
| Absorption coefficient, mm ⁻¹ | 1.436 | 1.328 |
| <i>F</i> ₀₀₀ | 1154 | 2436 |
| Crystal size, mm | 0.43 × 0.28 × 0.21 | 0.30 × 0.25 × 0.20 |
| θ range for data collection, deg | 1.61–30.03 | 3.02–30.03 |
| Index ranges | -16 ≤ <i>h</i> ≤ 16 -18 ≤ <i>k</i> ≤ 18 -27 ≤ <i>l</i> ≤ 27 | -15 ≤ <i>h</i> ≤ 15 -38 ≤ <i>k</i> ≤ 38 -28 ≤ <i>l</i> ≤ 28 |
| Reflections collected | 36781 | 174348 |
| Unique / observed (<i>I</i> > 2 σ (<i>I</i>)) reflections | 16547 / 14934 | 17942 / 13518 |
| <i>R</i> _{int} | 0.0261 | 0.0626 |
| Completeness to θ , % | 99.8 | 99.8 |
| Data / restraints / parameters | 16547 / 92 / 689 | 17942 / 1364 / 972 |
| <i>S</i> (<i>F</i> ²) | 1.032 | 1.017 |
| Final <i>R</i> indices (<i>F</i> ² > 2 σ (<i>F</i> ²)) | <i>R</i> ₁ = 0.0266 <i>wR</i> ₂ = 0.0573 | <i>R</i> ₁ = 0.0313 <i>wR</i> ₂ = 0.0620 |
| <i>R</i> indices (all data) | <i>R</i> ₁ = 0.0319 <i>wR</i> ₂ = 0.0593 | <i>R</i> ₁ = 0.0544 <i>wR</i> ₂ = 0.0693 |
| Largest diff. peak and hole, e/Å ³ | 0.81 / -0.54 | 0.56 / -0.54 |

Table S2: Fitting of the Cole-Cole plots with a generalized Debye model under a zero dc field for **1**.

| T (K) | χ_s (cm ³ . mol ⁻¹) | χ_T (cm ³ . mol ⁻¹) | α |
|---------|---|---|----------|
| 2 | 0.8946 | 7.00336 | 0.29475 |
| 4 | 0.53619 | 3.73958 | 0.29746 |
| 6 | 0.40245 | 2.54412 | 0.28608 |
| 8 | 0.34036 | 1.93861 | 0.25528 |
| 10 | 0.2945 | 1.55602 | 0.21416 |
| 11 | 0.27602 | 1.41483 | 0.19213 |
| 12.6 | 0.25743 | 1.23083 | 0.14327 |
| 14.2 | 0.22599 | 1.09578 | 0.12419 |
| 15.8 | 0.20117 | 0.98596 | 0.10767 |
| 17.4 | 0.17413 | 0.92276 | 0.12018 |
| 19 | 0.16848 | 0.82108 | 0.06973 |
| 19.5 | 0.16208 | 0.79604 | 0.06638 |
| 20 | 0.15636 | 0.78071 | 0.06558 |
| 20.5 | 0.14498 | 0.75887 | 0.07035 |
| 21 | 0.15237 | 0.74355 | 0.05432 |
| 21.5 | 0.11857 | 0.73635 | 0.10482 |
| 22 | 0.13059 | 0.71198 | 0.06349 |
| 22.5 | 0.13859 | 0.68724 | 0.04775 |
| 23 | 0.13096 | 0.67976 | 0.05399 |

Table S3: Fitting of the Cole-Cole plots with a generalized Debye model under a zero dc field for **2**.

| T (K) | χ_s (cm ³ . mol ⁻¹) | χ_T (cm ³ . mol ⁻¹) | α |
|---------|---|---|----------|
| 2.00 | 0.61786 | 0.02901 | 0.42505 |
| 4.25 | 0.47754 | 0.01323 | 0.38148 |
| 6.50 | 0.31783 | 0.01499 | 0.41276 |
| 11.00 | 0.39731 | 0.01539 | 0.41496 |
| 12.00 | 0.40697 | 0.0254 | 0.41327 |
| 13.00 | 0.429 | 0.00748 | 0.36118 |
| 14.00 | 0.40414 | 0.02144 | 0.36691 |
| 15.00 | 0.40564 | 0.00635 | 0.3363 |
| 16.00 | 0.39338 | 0.00469 | 0.32208 |
| 17.00 | 0.38108 | 0.00574 | 0.31179 |
| 18.00 | 0.33078 | 0.04602 | 0.37196 |
| 18.50 | 0.33029 | 0.00681 | 0.3413 |

| | | | |
|-------|---------|---------|---------|
| 19.20 | 0.2995 | 0.00738 | 0.37111 |
| 19.90 | 0.28751 | 0.00924 | 0.35876 |

Table S4: Fit parameters of the field dependence of the relaxation time for **1** using Eq.1 .

| Compound | $\Delta(cm^{-1})$ | $\tau_0 (s)$ | n | $C (s^{-1}.K^{-n})$ | $\tau_{QTM} (ms)$ |
|---|-------------------|-----------------------------|---------------|----------------------------------|-------------------|
| 1 (0 Oe) Obtained without considering the log normal distribution | 22 ± 6 | $(3 \pm 2) \times 10^{-10}$ | 5.7* | $(1.03 \pm 0.03) \times 10^{-4}$ | 1.56 ± 0.09 |
| 1 (0 Oe) With the Log normal distribution | 203 ± 1453 | $10^{-10(20)}$ | 1.03 ± 11 | $10^{-2(20)}$ | $10^{-3(0.3)}$ |

*Fixed parameter

Table S5: Fit parameters of the field dependence of the relaxation time for **1-3** using the extracted τ values.

| Compound | n | $C (s^{-1}.K^{-m})$ | $\tau_{QTM} (s)$ |
|--------------------|-----------------|--------------------------------|----------------------|
| 1 (0 Oe) | 5.04 ± 0.09 | 0.0009 ± 0.0002 | - |
| 1 (1000 Oe) | 5.74 ± 0.08 | 0.00006 ± 0.00002 | 0.00144 ± 0.0006 |
| 2 (0 Oe) | 8.3* | $(9.5 \pm 0.7) \times 10^{-9}$ | 0.00144 ± 0.0006 |
| 3 (0 Oe) | 4.2 ± 0.2 | 0.02 ± 0.01 | 0.0012 ± 0.0001 |
| 3 (3500 Oe) | 6.3 ± 0.3 | 0.01 ± 0.00001 | - |

*Fixed parameter

Table S6: Fit parameters of the field dependence of the relaxation time for **1** and **2**.

| Compound | $D (s^{-1}K^{-1}Oe^{-4})$ | $B_1 (s^{-1})$ | $B_2 (Oe^{-2})$ | K |
|-----------------|---------------------------|----------------|-----------------------|--------|
| 1 (15 K) | 8.3×10^{-15} | 1169.9 | 1.06×10^{-4} | 399.08 |
| 2 (10 K) | 2.1×10^{-15} | 1527.21 | 1.4×10^{-4} | 56.24 |

Table S7: Fitting of the Cole-Cole plots with a generalized Debye model under a 1000 Oe dc field for **1**.

| T (K) | χ_s (cm ³ . mol ⁻¹) | χ_T (cm ³ . mol ⁻¹) | α |
|---------|---|---|----------|
| 8 | 0.22814 | 1.92339 | 0.04864 |
| 10 | 0.18638 | 1.55864 | 0.05512 |
| 11 | 0.17713 | 1.42737 | 0.04033 |
| 12.6 | 0.15609 | 1.23843 | 0.05049 |
| 14.2 | 0.14409 | 1.08596 | 0.03572 |
| 15.8 | 0.13291 | 0.99804 | 0.04746 |
| 17.4 | 0.12486 | 0.90834 | 0.04728 |
| 19 | 0.11506 | 0.83443 | 0.05724 |
| 19.5 | 0.11178 | 0.81322 | 0.06167 |
| 20 | 0.11379 | 0.79386 | 0.05655 |
| 20.5 | 0.1117 | 0.77002 | 0.05306 |
| 21 | 0.10464 | 0.7574 | 0.06802 |
| 21.5 | 0.10275 | 0.74146 | 0.07151 |
| 22 | 0.11139 | 0.7243 | 0.05928 |
| 22.5 | 0.10454 | 0.70881 | 0.07011 |
| 23 | 0.10426 | 0.69394 | 0.06457 |
| 23.5 | 0.10167 | 0.67913 | 0.06447 |

Table S8. Fitting of the Cole-Cole plots under a 2000 Oe dc field for **2**.

| T (K) | χ_{Tot} (cm ³ . mol ⁻¹) | $\Delta\chi_1$ (cm ³ . mol ⁻¹) | α_1 | $\Delta\chi_2$ (cm ³ . mol ⁻¹) | α_2 |
|---------|---|---|------------|---|------------|
| 8 | 0.23 | 0.395 | 0.199 | 0.708 | 0.437 |
| 9.5 | 0.00536 | 0.346 | 0.231 | 0.794 | 0.504 |
| 11 | 0.00506 | 0.319 | 0.245 | 0.674 | 0.477 |
| 12 | 0.0219 | 0.323 | 0.268 | 0.571 | 0.357 |
| 13 | 0.0368 | 0.288 | 0.26 | 0.527 | 0.405 |
| 14 | 0.347 | 0.293 | 0.276 | 0.156 | 0.0775 |
| 15 | 0.281 | 0.274 | 0.281 | 0.192 | 0.118 |
| 16 | 0.247 | 0.11 | 0.0857 | 0.356 | 0.602 |
| 17 | 5.86E-7 | 0.131 | 0.149 | 0.545 | 0.673 |
| 18 | 7.94E-13 | 0.131 | 0.19 | 0.508 | 0.658 |
| 18.5 | 2.7E-13 | 0.104 | 0.154 | 0.517 | 0.607 |
| 19.2 | 4.8E-13 | 0.129 | 0.205 | 0.473 | 0.663 |
| 19.9 | 3.83E-12 | 0.0739 | 0.116 | 0.507 | 0.613 |

| | | | | | |
|------|----------|-------|-------|-------|-------|
| 20.6 | 7.39E-11 | 0.22 | 0.361 | 0.336 | 0.472 |
| 21.3 | 6.86E-11 | 0.186 | 0.35 | 0.357 | 0.623 |

References

- 1 N. F. Chilton, D. Collison, E. J. L. McInnes, R. E. P. Winpenney and A. Soncini, *Nat. Commun.*, 2013, **4**, 2551.