Supporting Information For:

Slow Magnetic Relaxation in Homoleptic Trispyrazolylborate Complexes of Neodymium(III) and Uranium(III)

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Synthesis.

The molecular compound $NdTp_3$ was prepared according to a previously published procedure by reaction of $NdCl_3$ with KTp in water,[1] and UTp_3 was prepared using a modification of a published procedure.[2] Under a dry nitrogen atmosphere, a THF solution (8 mL) of UI_3 (200 mg, 0.323 mmol) was added to a stirring THF solution (8 mL) of KTp (244.4 mg, 0.970 mmol). After stirring for 12 h, a grayish brown solid was filtered from a clear, pale yellow solution. The solid was washed with dry MeOH (3 x 10 mL) and isolated as a dark brown solid (228 mg, 81%). Dilute $NdTp_3$ samples were prepared by adding a stoichiometric amount of KTp to a vigorously stirring solution of $LaTp_3$ and $NdTp_3$ in the desired molar ratio.

Magnetic Measurements.

The magnetic samples of $NdTp_3$ and UTp_3 were prepared by adding samples to 7 mm quartz tubes. Sufficient liquid eicosane (at 40 °C) was added to saturate and cover the samples to prevent crystallite torqueing and provide good thermal contact between the sample and the bath. Tubes were fitted with sealable adapters, evacuated on a Schlenk line, and flame sealed under vacuum.

Magnetic susceptibility measurements were collected using a Quantum Design MPMS2 SQUID magnetometer. DC susceptibility data measurements were performed at temperatures ranging from 2 to 300 K, using applied fields of 1000, 5000, and 10,000 Oe. Ac magnetic susceptibility data were collected using a 4 Oe switching field. All data were corrected for diamagnetic contributions from the eicosane and core diamagnetism estimated using Pascal's constants of χ_D = –0.0004 emu/mol.

Cole-Cole plots were fitted using formulae describing χ' and χ'' in terms of frequency, constant temperature susceptibility (χ_T), adiabatic susceptibility (χ_S), relaxation time (τ), and a variable representing the distribution of relaxation times (α).[3]

Table S1 | Table of constant temperature susceptibility (χ_T), adiabatic susceptibility (χ_S), relaxation time (τ), and a variable representing the distribution of relaxation times (α) for various dilutions of $NdTp_3$.

100 mol % $NdTp_3$				
Temperature(K)	**X_T (emu/mol)**	**X_s (emu/mol)**	**α**	**τ (s)**
1.8	0.2136	0.0335	0.035	0.000402(2)
1.9	0.2074	0.0319	0.034	0.000363(2)
2	0.2007	0.0307	0.032	0.000328(3)
2.1	0.1936	0.0292	0.031	0.000295(2)
2.2	0.1884	0.0279	0.032	0.000273(3)
2.3	0.1814	0.0272	0.029	0.000247(2)
2.4	0.1748	0.0266	0.027	0.000225(2)
59 mol % $NdTp_3$				
Temperature(K)	**X_T (emu/mol)**	**X_s (emu/mol)**	**α**	**τ (s)**
1.8	0.1404	0.0187	0.036	0.00082(1)
1.9	0.1348	0.0180	0.034	0.000727(8)
2	0.1294	0.0173	0.033	0.000645(6)
2.1	0.1242	0.0163	0.034	0.000572(5)
2.2	0.1207	0.0157	0.036	0.000527(3)
2.3	0.1154	0.0151	0.034	0.000468(4)
2.4	0.1108	0.0146	0.034	0.000418(5)
15 mol % $NdTp_3$				
Temperature(K)	**X_T (emu/mol)**	**X_s (emu/mol)**	**α**	**τ (s)**
1.8	0.2048	0.0179	0.092	0.00174(2)
1.9	0.1962	0.0159	0.097	0.00155(2)
2	0.1870	0.0159	0.095	0.00140(1)
2.1	0.1787	0.0143	0.099	0.00124(2)
2.2	0.1731	0.0147	0.097	0.001156(7)
2.3	0.1646	0.0128	0.101	0.00101(2)
2.4	0.1585	0.0118	0.108	0.00090(2)
3.8 mol % $NdTp_3$				
Temperature(K)	**X_T (emu/mol)**	**X_s (emu/mol)**	**α**	**τ (s)**
1.8	0.15	0.00	0.22	0.0053(2)
1.9	0.14	0.00	0.23	0.0044(3)
2	0.13	0.00	0.22	0.0039(2)
2.1	0.13	0.00	0.21	0.0034(2)
2.2	0.12	0.00	0.22	0.0031(2)
2.3	0.12	0.00	0.23	0.0028(3)

Table S2 | Table of constant temperature susceptibility (χ_T), adiabatic susceptibility (χ_S), relaxation time (τ), and a variable representing the distribution of relaxation times (α) for UTp_3.

UTp_3				
Temperature(K)	**X_T (emu/mol)**	**X_s (emu/mol)**	**α**	**τ (s)**
1.9	0.79	0.16	0.03	0.001245(3)
2	0.75	0.14	0.04	0.001077(4)
2.1	0.72	0.13	0.04	0.000949(3)
2.3	0.66	0.13	0.03	0.000765(2)
2.4	0.63	0.12	0.03	0.000675(2)

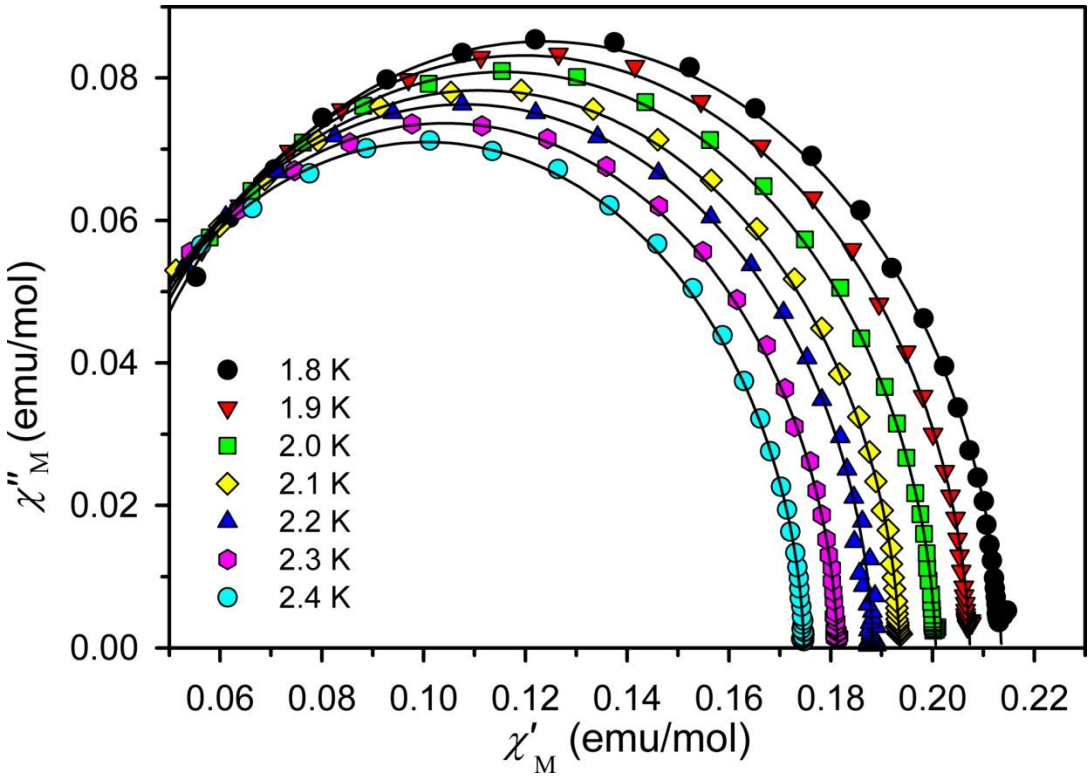

Figure S1. Cole-Cole (Argand) plots for ac susceptibility data collected on $NdTp_3$ at 100 Oe and varying temperatures. Experimental data points are represented by symbols with the black line representing a fit to a generalized Debye equation.[3]

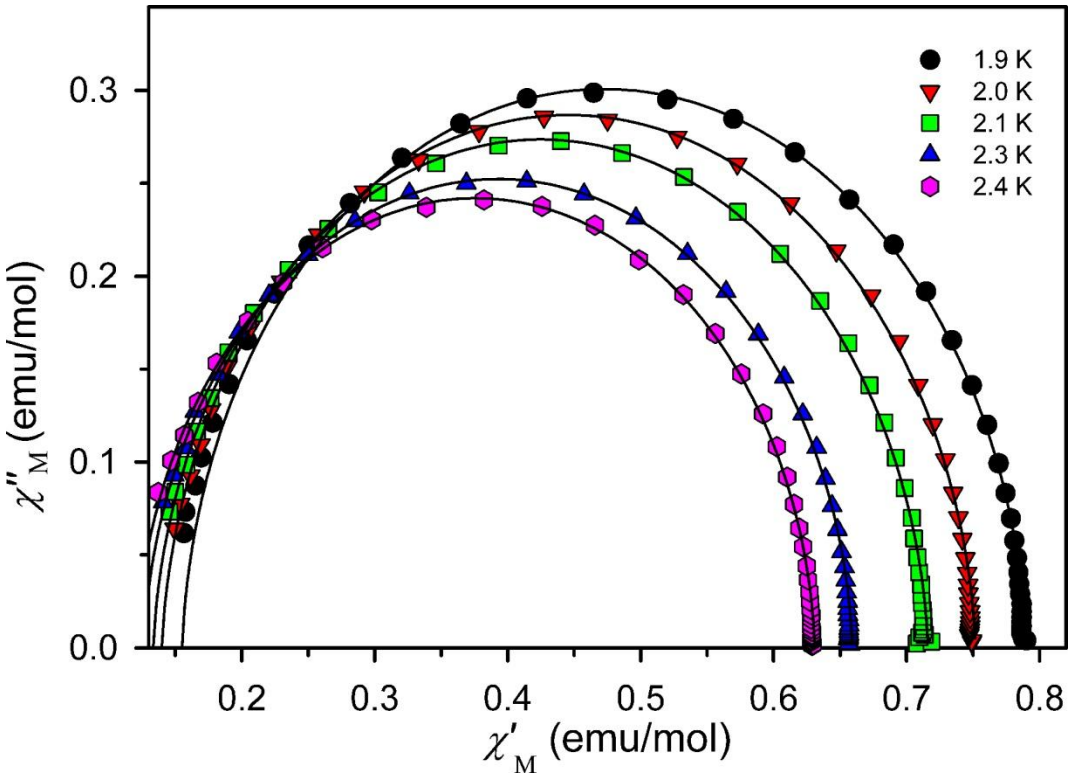

Figure S2. Cole-Cole (Argand) plots for ac susceptibility data collected on UTp_3 at 100 Oe and varying temperatures. Experimental data points are represented by symbols with the black line representing a fit to a generalized Debye equation.[3]

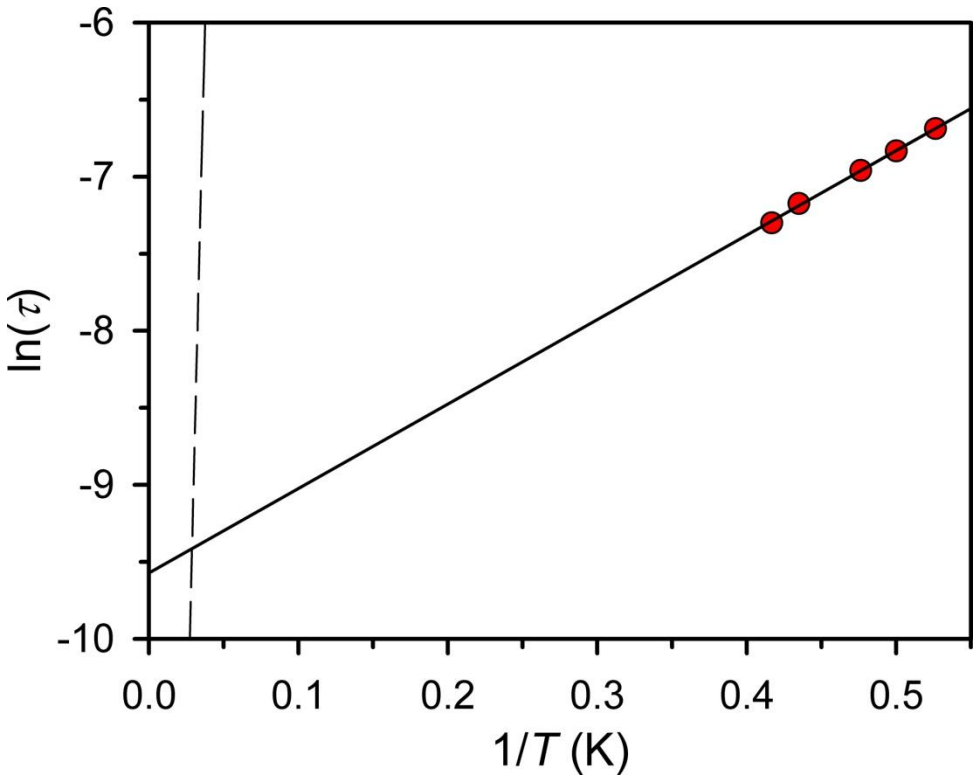

Figure S3. Plot of inverse temperature vs. the natural log of the magnetic relaxation time for UTp_3. The solid black line represents an Arrhenius fit with parameters U_{eff} = 3.81(8) cm^{-1} and τ_0 = 7.0(4) x 10^{-5} s The dashed line represents theoretical Arrhenius behavior (U_{eff} = 267 cm^{-1}; τ_0 = 10^{-9} s) predicted by the electronic structure.

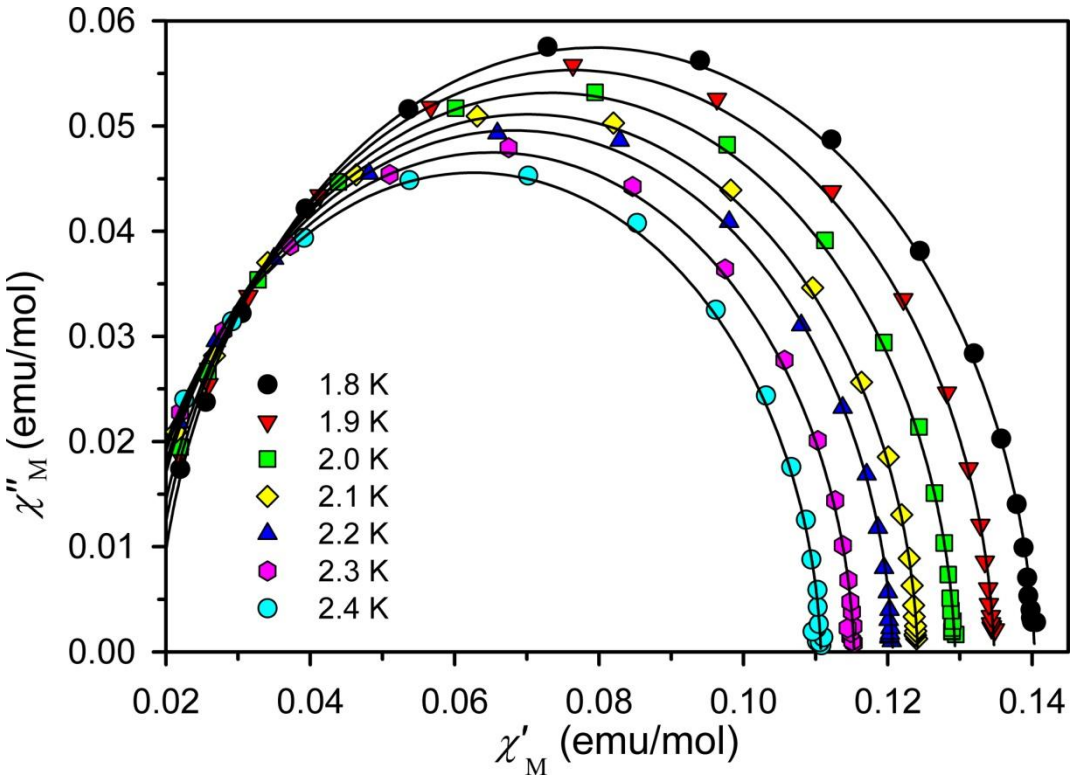

Figure S4. Cole-Cole (Argand) plots for ac susceptibility data collected for a 59 mol. % $NdTp_3$ sample at 100 Oe and varying temperatures. Experimental data points are represented by symbols with the black line representing a fit to a generalized Debye equation.[3]

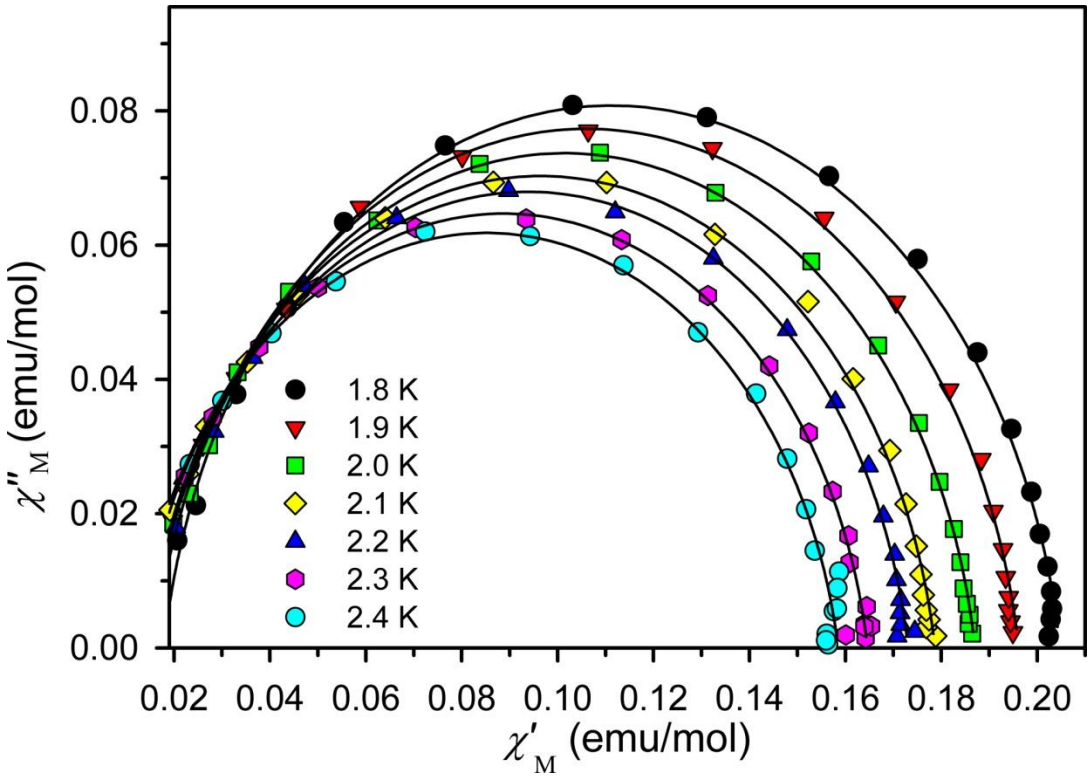

Figure S5. Cole-Cole (Argand) plots for ac susceptibility data collected for a 15 mol. % $NdTp_3$ sample at 100 Oe and varying temperatures. Experimental data points are represented by symbols with the black line representing a fit to a generalized Debye equation.[3]

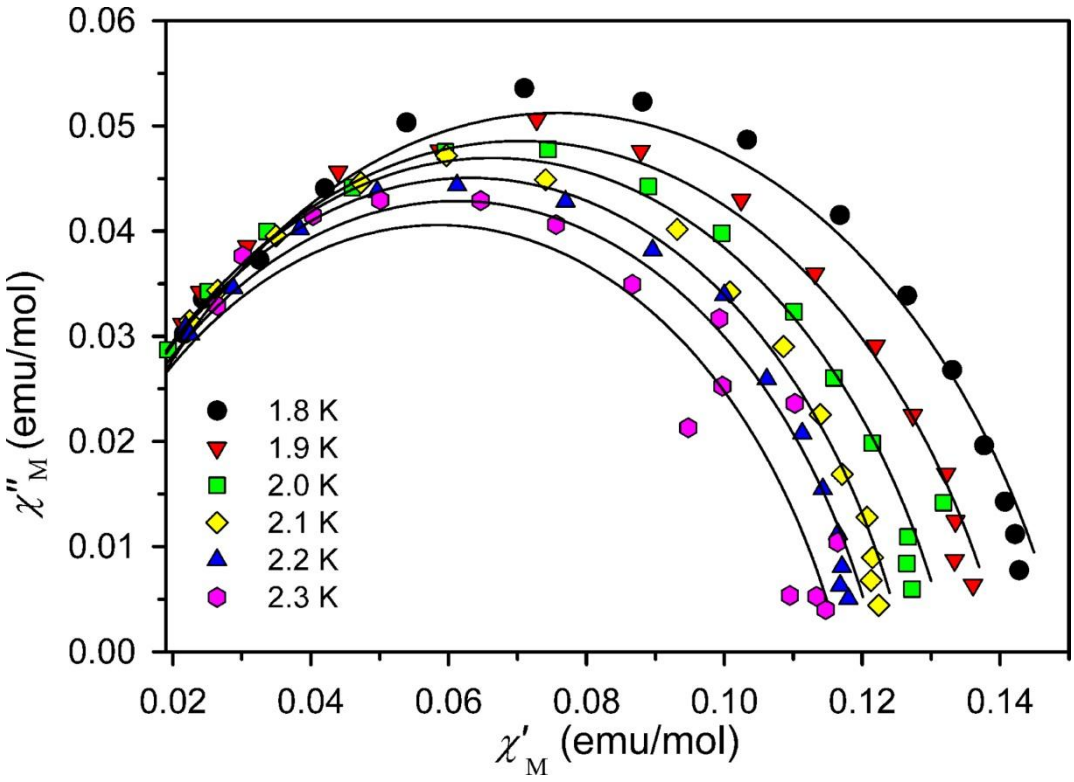

Figure S6. Cole-Cole (Argand) plots for ac susceptibility data collected for a 3.8 mol. % $NdTp_3$ sample at 100 Oe and varying temperatures. Experimental data points are represented by symbols with the black line representing a fit to a generalized Debye equation.[3]

(1) Apostolidis, C.; Rebizant, J.; Kanellakopulos, B.; von Ammon, R.; Dornberger, E.; Mlaller, J.; Powietzka, B., Nuber, B. *Polyhedron* **1997**, *16*, 1057.

(2) Apostolidis, C.; Morgenstern, A.; Rebizant, J.; Kanellakopulos, B.; Walter, O.; Powietzka, B.; Karbowiak, M.; Reddmann, H.; Amberger, H.-D. *Z. Anorg. Allg. Chem.* **2010**, *636*, 201.

(3) Gatteschi, D., Sessoli, R., Villain, J. *Molecular Nanomagnets*, Oxford University Press, Oxford, **2006**.