Supplementary Information

Table S1. Crystal data and structure refinement for **1-5**.

Identification code	**1**	**2**	**3**	**4**	**5**
Empirical formula	$C_{27}H_{25}DyN_4O_6$	$C_{27}H_{25}HoN_4O_6$	$C_{27}H_{25}ErN_4O_6$	$C_{27}H_{25}N_4O_6Tm$	$C_{27}H_{25}N_4O_6Yb$
Formula weight	664.01	666.44	668.77	670.44	674.55
Temperature, K	150(2)	150(2)	150(2)	120(2)	150(2)
Wavelength, Å	0.71073	0.71073	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$
a, Å	12.9262(5)	12.9230(6)	12.9232(5)	12.8785(6)	12.9137(3)
b, Å	14.8199(5)	14.8501(6)	14.8811(6)	14.8515(6)	14.9039(4)
c, Å	13.7408(6)	13.7212(6)	13.7088(5)	13.6469(6)	13.6530(3)
β, °	102.1743(11)	102.1890(10)	102.2980(12)	102.2770(10)	102.4320(10)
Volume, $Å^3$	2573.06(17)	2573.84(19)	2575.86(17)	2550.48(19)	2566.10(11)
Z	4	4	4	4	4
D (calc), Mg/m^3	1.714	1.720	1.724	1.746	1.746
μ, mm^{-1}	2.954	3.124	3.308	3.529	3.694
F(000)	1316	1320	1324	1328	1332
Crystal size, mm	0.28 x 0.28 x 0.12	0.16 x 0.10 x 0.08	0.16 x 0.10 x 0.08	0.20 x 0.20 x 0.12	0.40 x 0.24 x 0.20
θ range, °	2.046, 27.100	2.046, 31.539	2.046, 29.572	2.053, 34.360	2.049, 27.101
Index ranges	-16<=h<=14	-19<=h<=18	-15<=h<=17	-19<=h<=18	-16<=h<=14
	-18<=k<=17	-21<=k<=21	-20<=k<=20	-22<=k<=21	-19<=k<=19
	-17<=l<=17	-20<=l<=19	-19<=l<=19	-20<=l<=19	-16<=l<=17
Reflections collected	22450	37448	35036	46111	26681
Independent reflections, Rint	5662, 0.0829	8533, 0.0290	7219, 0.0374	9636, 0.0305	5649, 0.0238
Completeness to θ = 25.242°	99.8 %	99.8 %	100.0 %	99.7 %	99.7 %
Absorption correction	Semi-empirical from equivalents	Semi-empirical from equivalents	Semi-empirical from equivalents	Semi-empirical from equivalents	Semi-empirical from equivalents
Max,. min. transmission	1, 0.9103	0.267, 0.2169	0.2671, 0.1815	0.7468, 0.5185	0.7461, 0.5077
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Data / restraints / parameters	5662 / 0 / 343	8533 / 0 / 347	7219 / 0 / 347	9636 / 0 / 348	5649 / 0 / 347
Goodness-of-fit	1.173	1.012	1.038	1.026	0.996
R1, wR2 [I>2sigma(I)]	0.0299, 0.0715	0.0233, 0.0551	0.0296, 0.0571	0.0199, 0.0444	0.0222, 0.0509
R1, wR2 (all data)	0.0325, 0.0726	0.0305, 0.0580	0.0402, 0.0603	0.0265, 0.0465	0.0253, 0.0523
Largest diff. peak and hole, $e.Å^{-3}$	2.264, -1.751	1.401, -0.811	2.343, -0.915	0.612, -0.752	2.453, -1.142
CCDC number	2365586	2365587	2365588	2365589	2365590

Table 2. Selected bond lengths [Å] for **1**-**5**.

	1 (Dy)	**2** (Ho)	**3** (Er)	**4** (Tm)	**5** (Yb)
Ln(1)-O(1)	2.314(2)	2.2960(15)	2.290(2)	2.2730(11)	2.270(2)
Ln(1)-O(3)	2.330(2)	2.3277(14)	2.3221(19)	2.3033(11)	2.2923(19)
Ln(1)-O(4)	2.297(2)	2.2857(14)	2.274(2)	2.2635(11)	2.2552(19)
Ln(1)-O(5)	2.349(2)	2.3452(15)	2.333(2)	2.3183(11)	2.305(2)
Ln(1)-O(6)	2.292(2)	2.2858(15)	2.277(2)	2.2634(11)	2.252(2)
Ln(1)-N(1)	2.545(3)	2.5281(18)	2.519(3)	2.5028(14)	2.502(2)
Ln(1)-N(3)	2.554(3)	2.5370(17)	2.523(2)	2.5104(13)	2.505(2)
Ln(1)-N(4)	2.523(3)	2.5074(16)	2.495(2)	2.4808(12)	2.478(2)

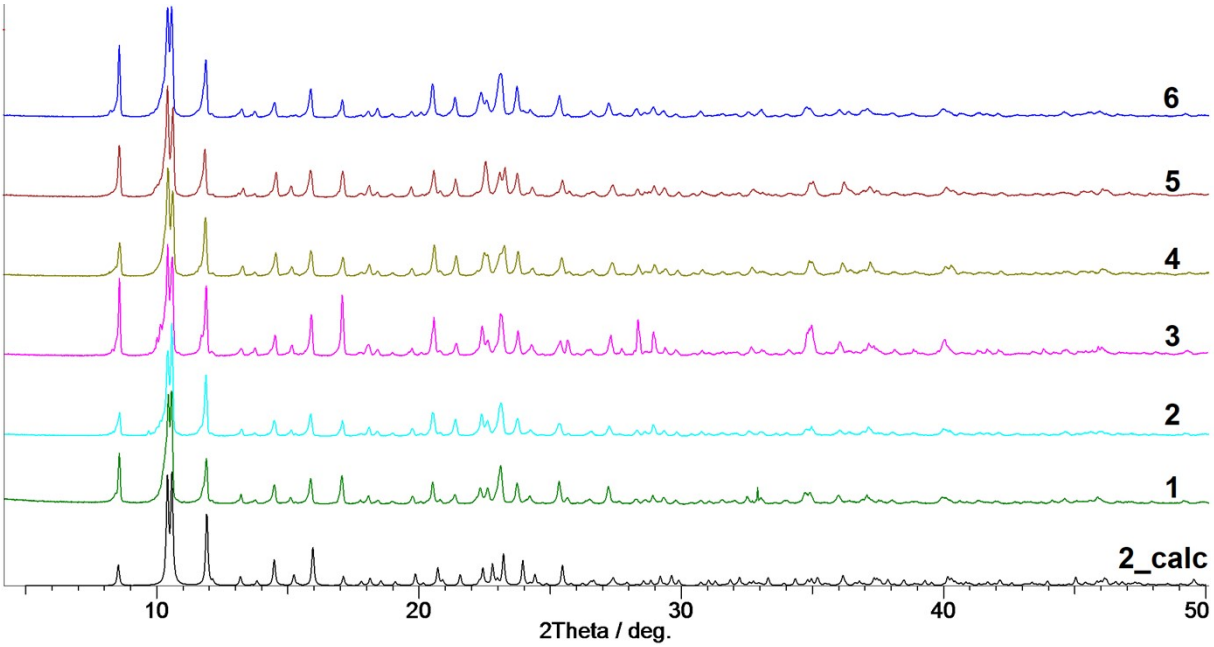


Fig. S1. Powder XRD patterns of samples isolated from the syntheses of **1**-**6**compared with the calculated pattern of **2** structure.

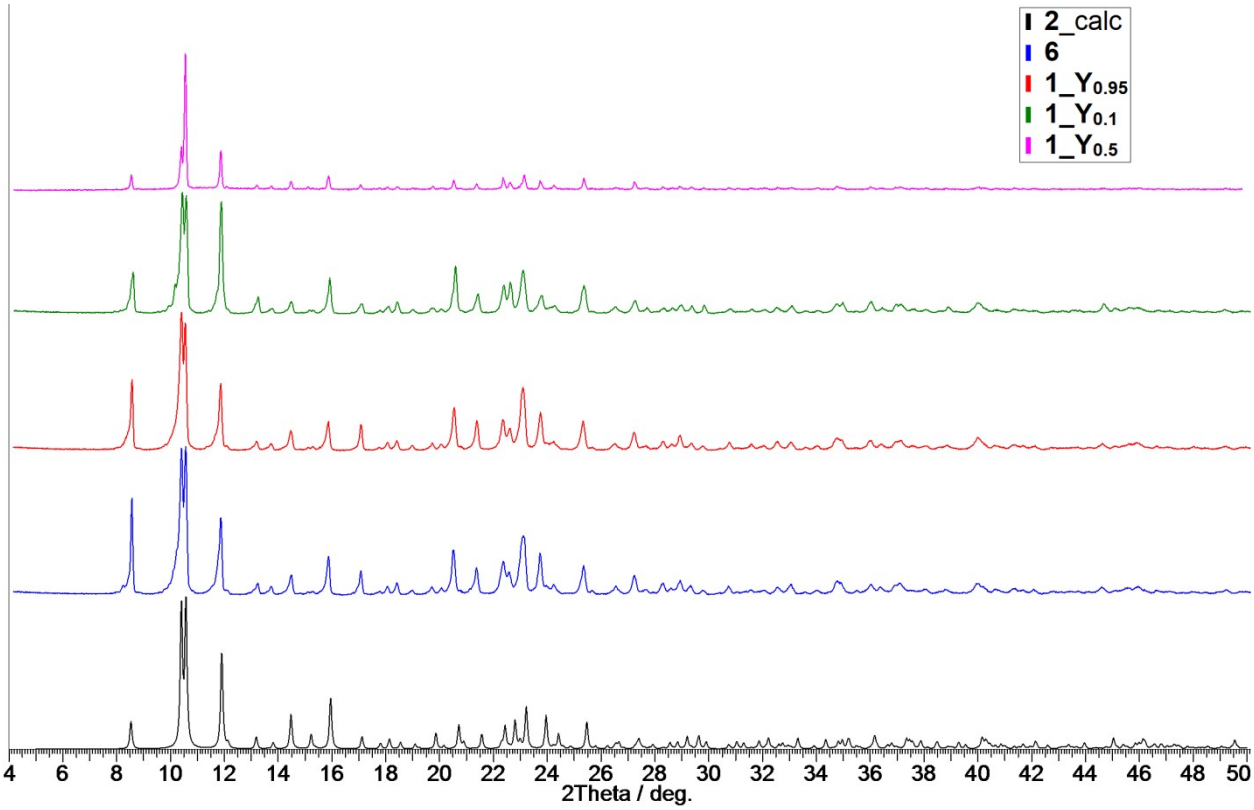


Fig. S2. Powder XRD patterns of Y-diluted samples compared with powder pattern of complex **6**(Y) and calculated pattern of **2**structure.

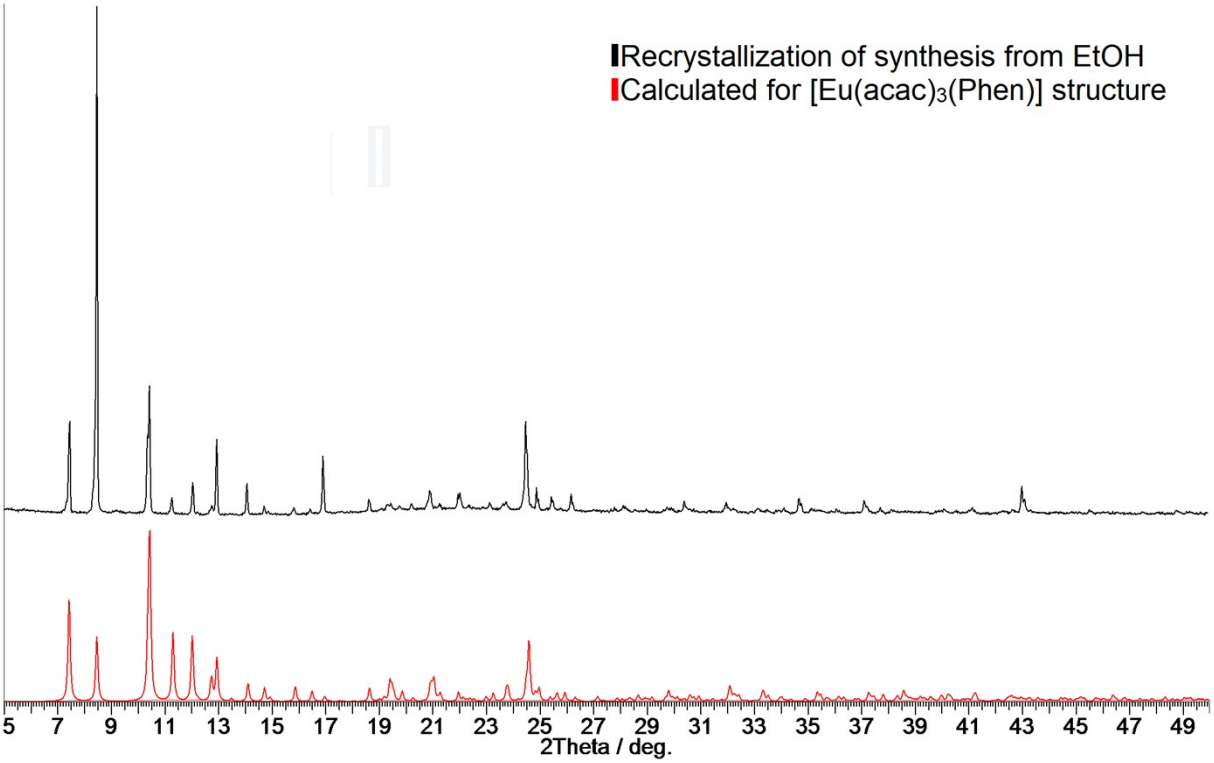


Fig. S3. Powder XRD patterns of product isolated upon the ethanolic solution of the product of interaction between $Dy(acac)_3 \cdot 3H_2O$, PyrCOOH and $Phen \cdot H_2O$ in THF:EtOH:H_2O (11:11:3) mixture compared with the calculated pattern of $[Eu(acac)_3(Phen)]$ structure.

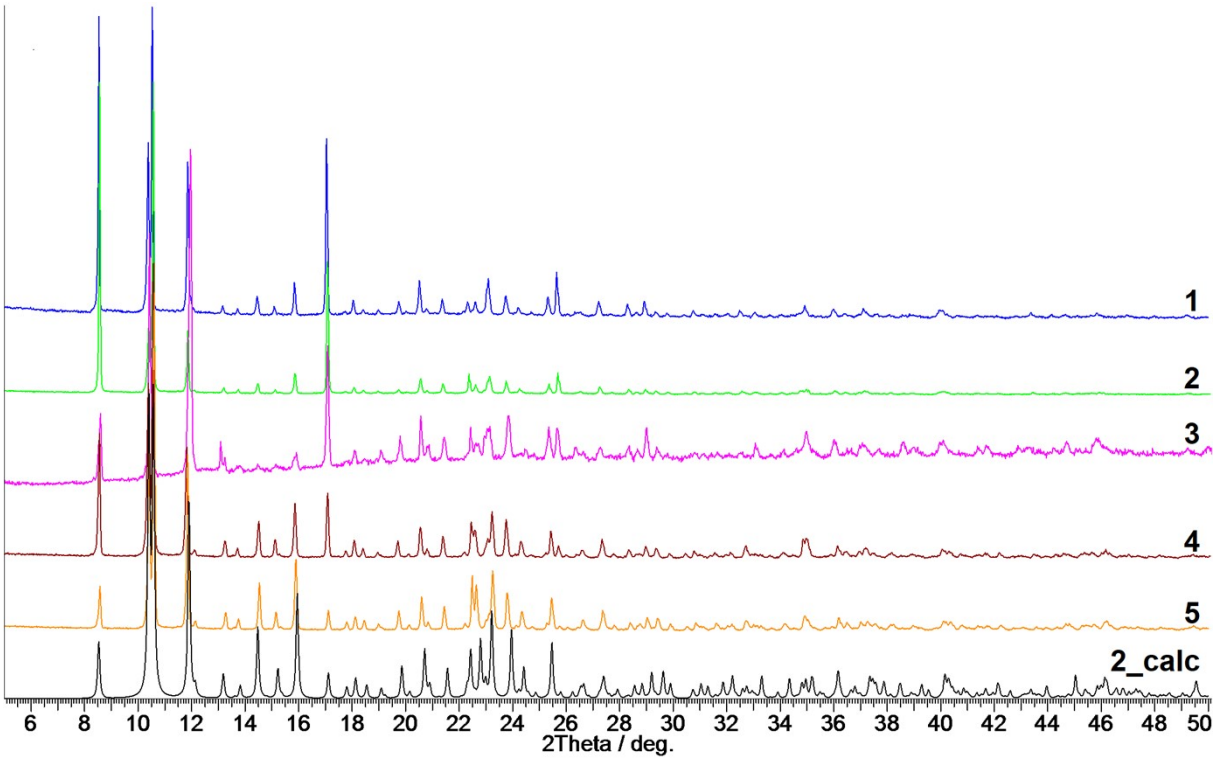


Fig. S4. Powder XRD patterns of samples isolated from the syntheses of **1**-**5***via* an alternative technique (involving the interaction of $Dy(acac)_3 \cdot 3H_2O$, PyrCOOH and $Phen \cdot H_2O$ in $THF:EtOH:H_2O$ (11:11:3) mixture with subsequent recrystallization from $^{n}PrOH$) compared with the calculated pattern of **2** structure.

Table S3. Continuous SHAPE measurements for **1** structure.

Structure	*SAPR-8*	*TDD-8*	*BTPR-8*
4	*0.982*	*1.867*	*2.631*

SAPR-8 Square antiprism
TDD-8 Triangular dodecahedron
BTPR-8 Biaugmented trigonal prism

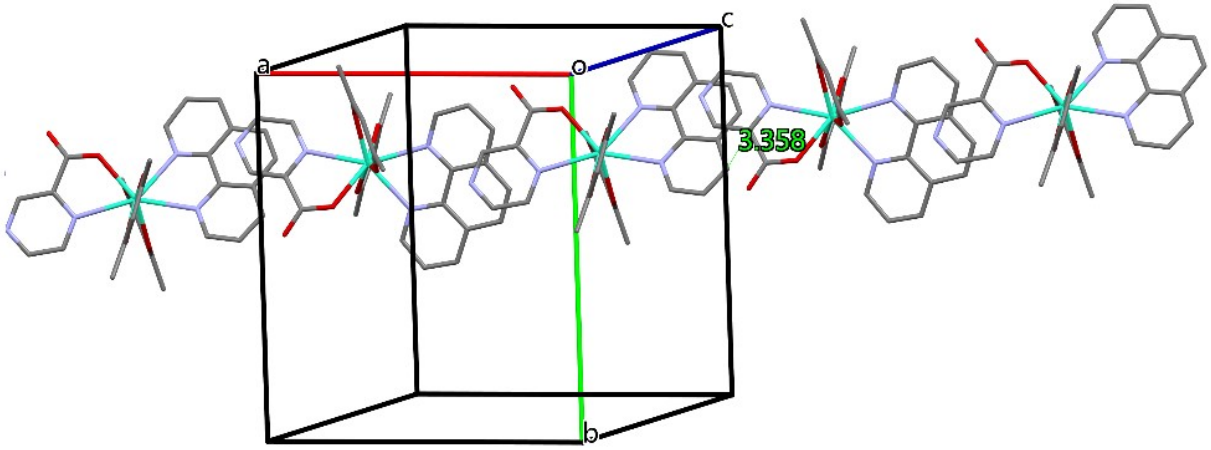


Fig. S5. Stacking interactions in the structure of **1**.

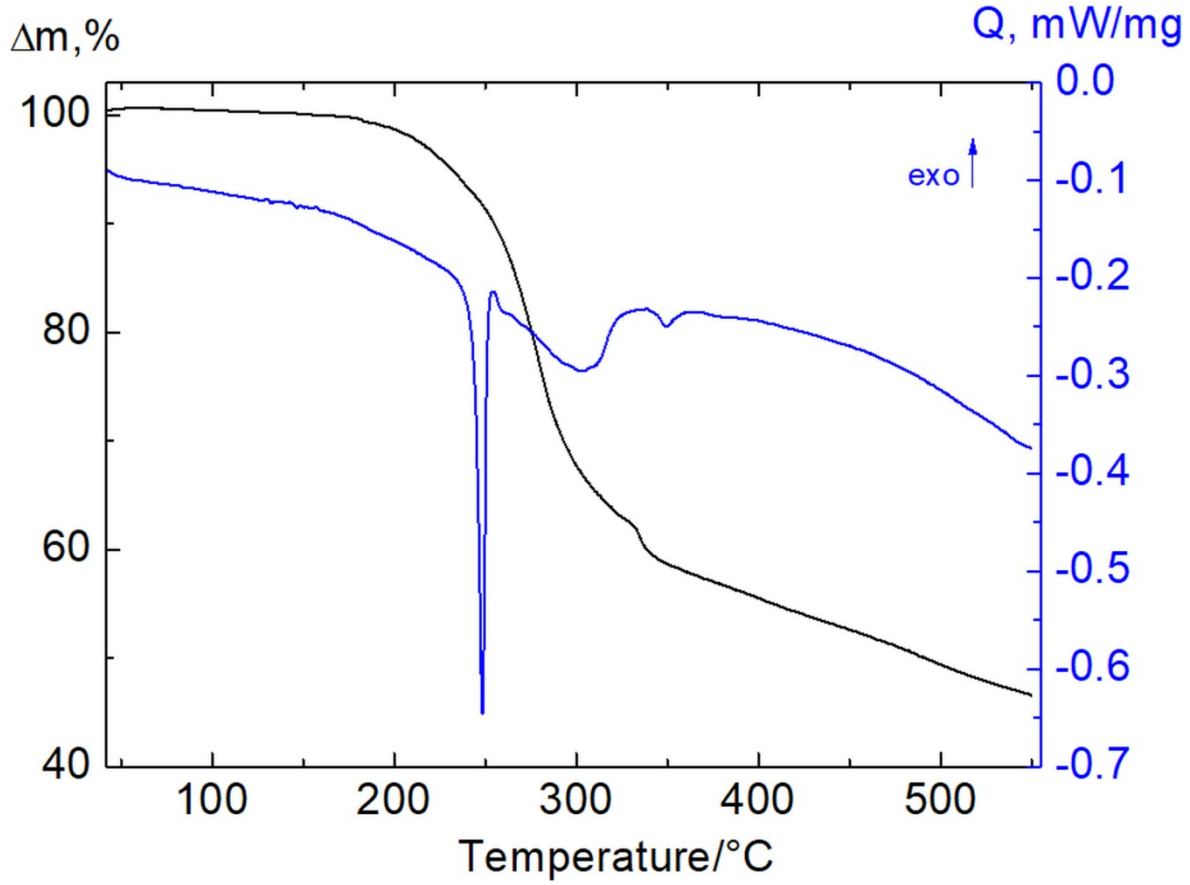


Fig. S6.TG (black) and DSC (blue) curves of complex **1** on heating under an Ar flow.

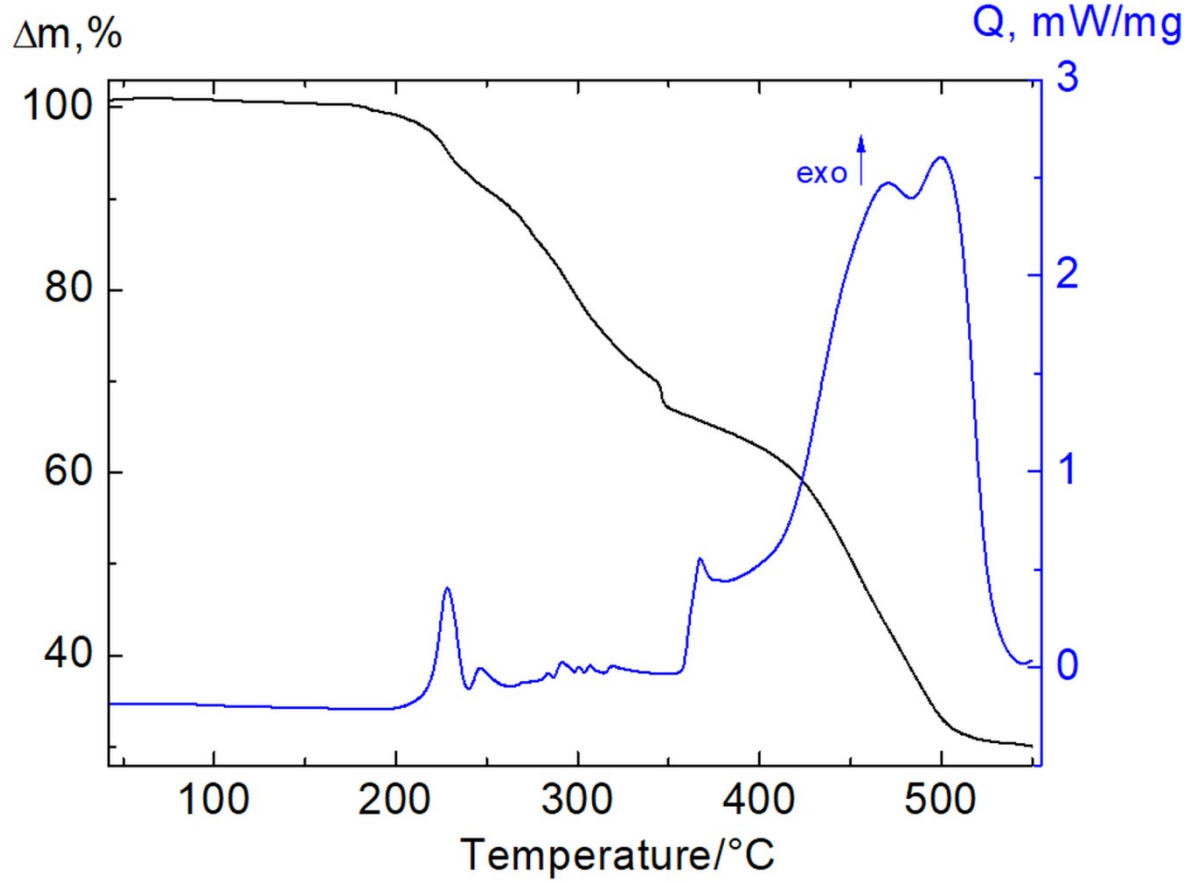


Fig. S7.TG (black) and DSC (blue) curves of complex **1** on heating under artificial air flow.

Table S4. Solubility of complex **5** in organic solvents. In each the case, 0.1 g of complex was taken. #Complex is virtually insoluble or much more than 10 ml of solvent was required for complete dissolution. *Solubility was probed at 80 °C rather than at corresponding boiling points.

Solvent	V_{Solv} at 25 °C, ml	V_{Solv} at *bp*, ml
$CHCl_3$	4	2
CH_2Cl_2	4	4
EtOH	>10#	6
EtOAc	>10#	>10#
THF	>10#	>10#
MeCN	>10#	10*

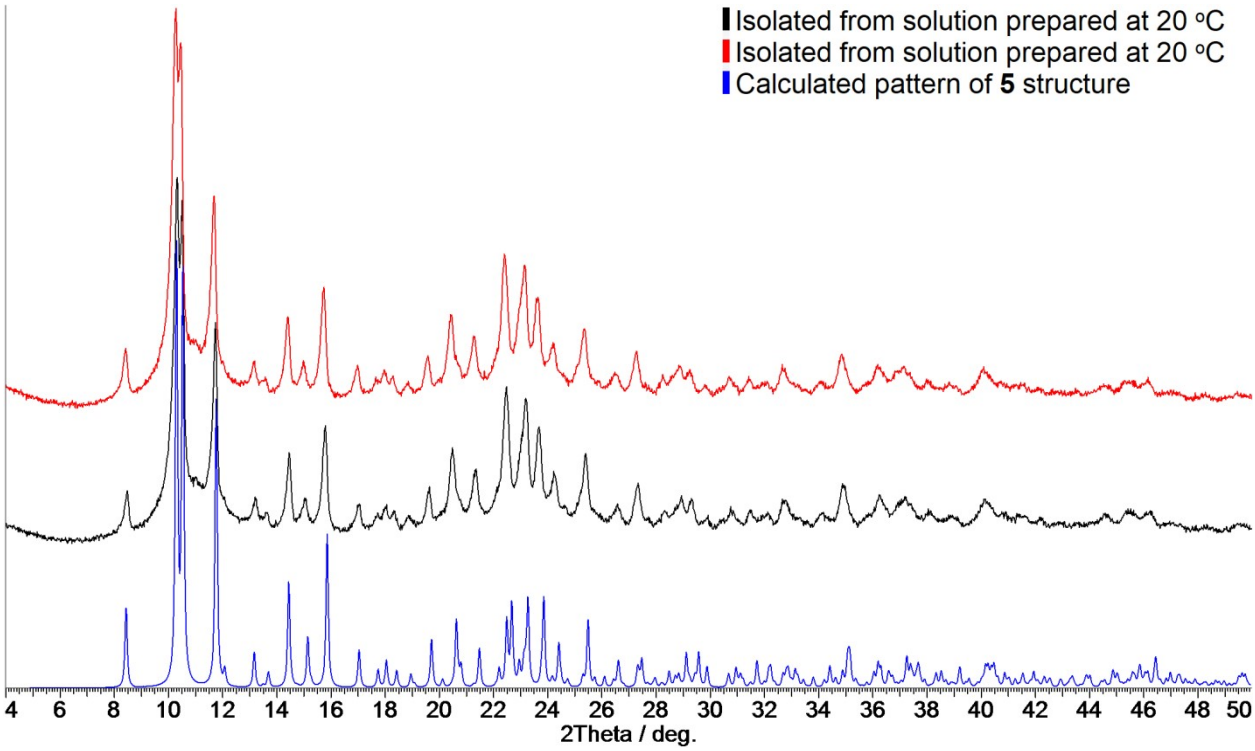


Fig. S8. Powder XRD patterns of the products isolated from the solutions of **5** in CH_2Cl_2 prepared at room temperature and at boiling point of CH_2Cl_2compared to calculated pattern of **5** structure.

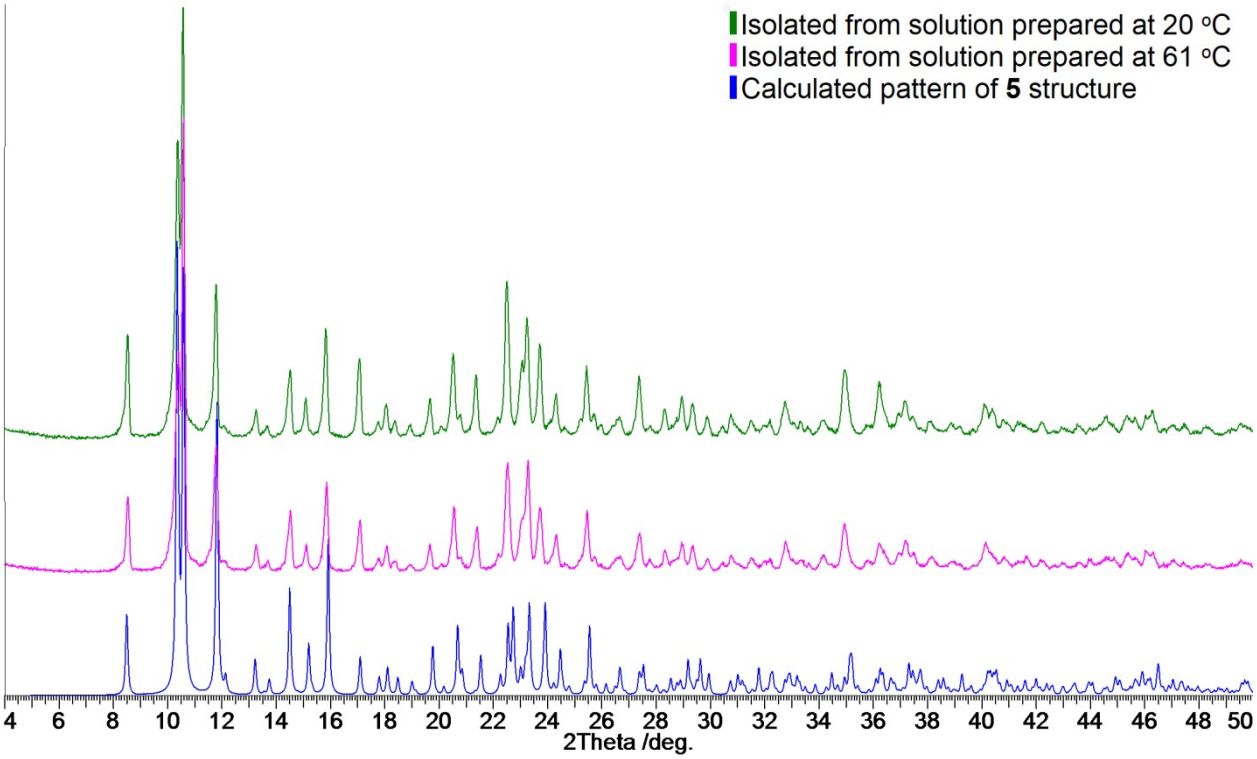
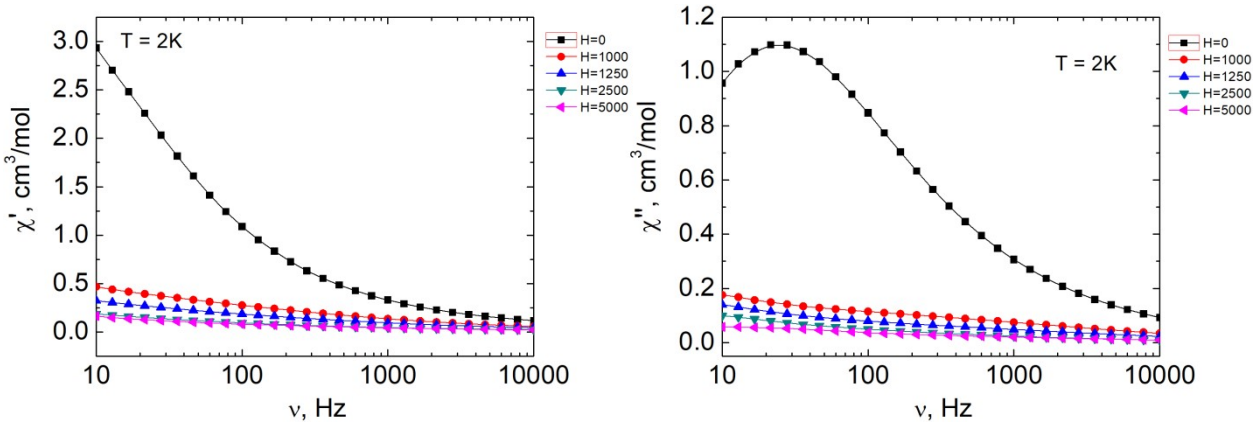


Fig. S9. Powder XRD patterns of the products isolated from the solutions of **5** in $CHCl_3$ prepared at room temperature and at boiling point of $CHCl_3$compared to calculated pattern of **5** structure.



FigS10. Frequency dependencies of real (left)and imaginary (right) components of dynamic magnetic susceptibility of complex **1**in various DC magnetic fields at 2 K. Solid lines are visual guides.

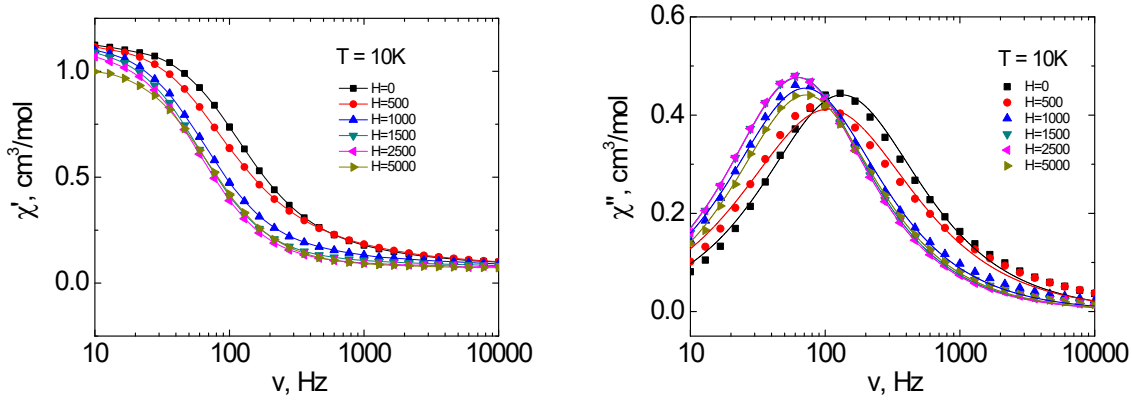
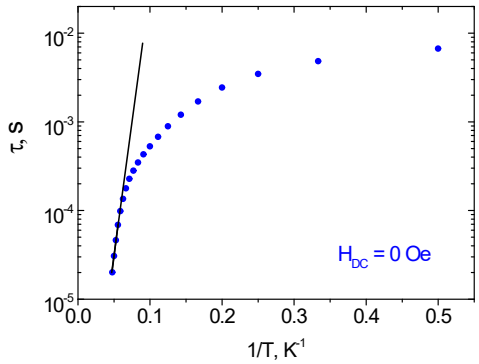
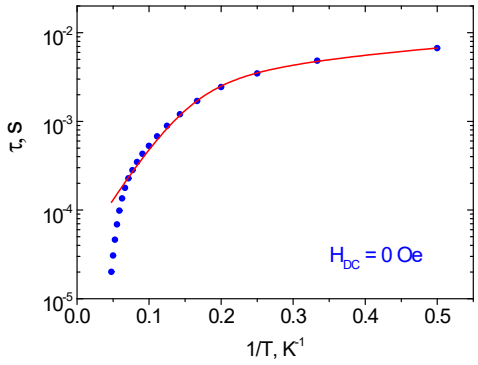
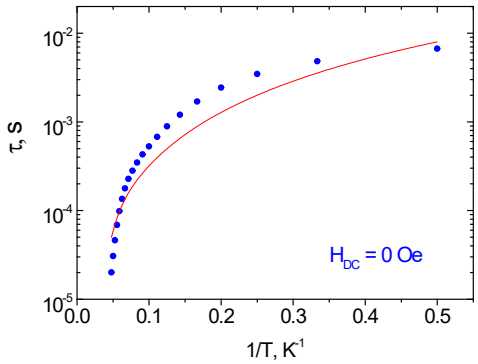
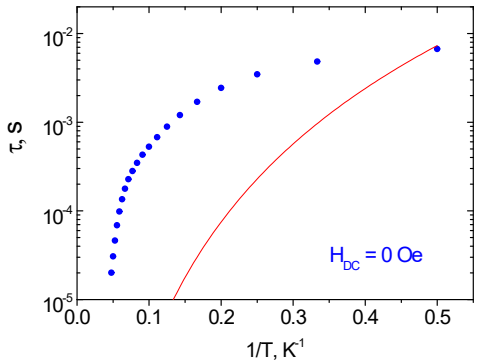
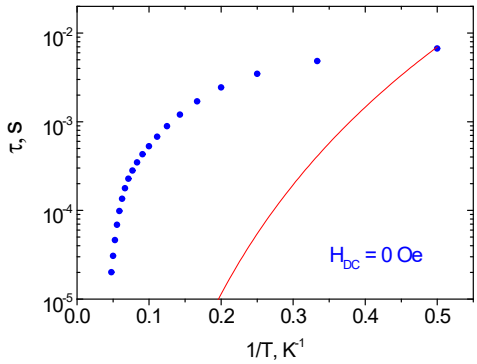
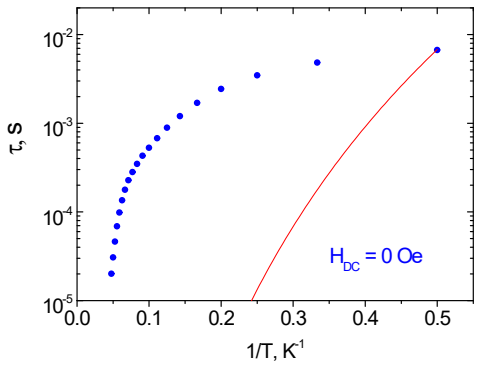
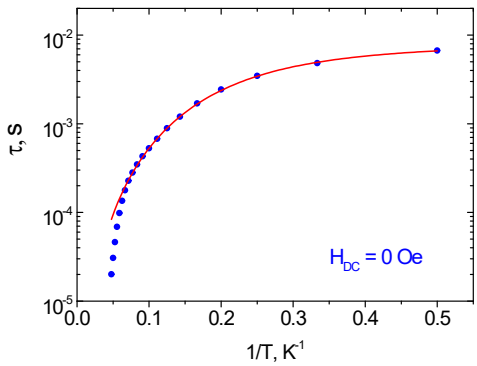
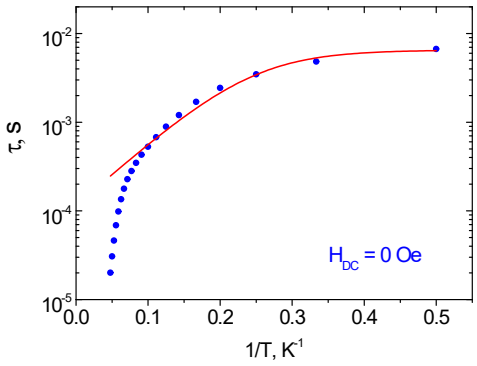


Fig S11. Frequency dependencies of real (left) and imaginary (right) components of dynamic magnetic susceptibility of complex **1**in various DC magnetic fields at 10 K. Solid lines are visual guides.

Table S5. Fitting of the τ *vs.* T dependences for complex **1** both under zero DC field (T = 2-21 K) and optimal DC field (H_{DC} = 1500 Oe, T = 8-22 K).Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color

Fitting	**Fit function,** **Best-fit parameters**
H_{DC} = 0 Oe	
	**Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)$ $\tau_0 = 1.78\cdot10^{-8}\pm5.8\cdot10^{-9}$s Δ_{eff}/k_B = 148.8 ± 6.0 K **R^2 = 0.99611**
	**Raman+Orbach** $\tau^{-1}=\tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = 83.45±0.8 $s^{-1}K^{-nRaman}$ n_{Raman} = 0.84±0.04 $\tau_0 = 3.44\cdot10^{-5}\pm 6.56\cdot10^{-6}$ s Δ_{eff}/k_B = 29.5 ± 1.6 K R^2 = 0.9986
	**Raman+Orbach** $\tau^{-1}=\tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ $\tau_0 = 1.3\cdot10^{-7}\pm 2.6\cdot10^{-6}$ s Δ_{eff}/k_B = 149 K (fixed) C_{Raman} = 31.2 ± 2.3 $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) R^2 = 0.86307

	**Raman+Orbach** $\tau^{-1}=\tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ $\tau_0 = 1 \pm 1.12\cdot10^{14}$ s Δ_{eff}/k_B = 149 K (fixed) C_{Raman} = 4.3± 0.8 $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) R^2 = 0.36767
	**Raman+Orbach** $\tau^{-1}=\tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ $\tau_0 = 1 \pm 1.21\cdot10^{17}$ s Δ_{eff}/k_B = 149 K (fixed) C_{Raman} = 1.12± 0.25 $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) R^2 = 0.26864
	**Raman+Orbach** $\tau^{-1}=\tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ τ_0 = 1 s Δ_{eff}/k_B = 149 K (fixed) C_{Raman} = 0.29± 0.07 $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) R^2 = 0.22878
	**Raman+QTM** $\tau^{-1}=B+C_{Raman}T^{nRaman}$ B = 122 ±2 s^{-1} C_{Raman} = 4.83±0.4 $s^{-1}K^{-nRaman}$ n_{Raman} = 2.56±0.04 **R^2 = 0.9994**



Orbach+QTM

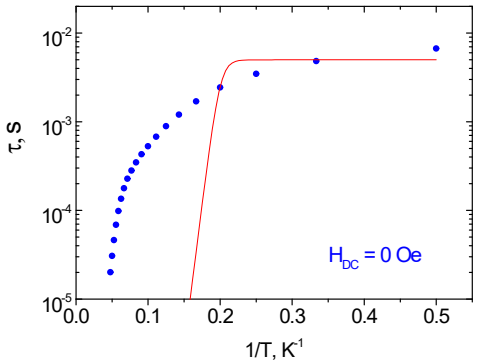
$\tau^{-1}=\tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B$

B = 153.7 ±5.4 s^{-1}

$\tau_0 = 1.17\cdot10^{-4}\pm 2.45\cdot10^{-5}$ s

Δ_{eff}/k_B = 16.5 ± 1.1 K

R^2 = 0.9855



Orbach+QTM

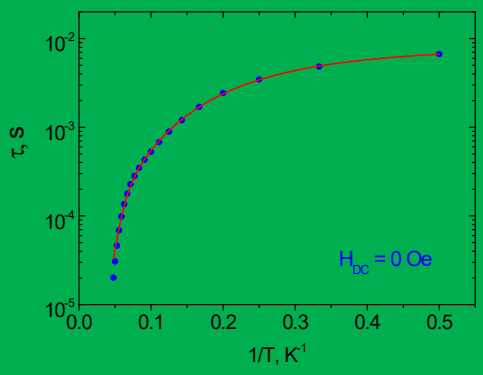
$\tau^{-1}=\tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B$

B = 200.4± 18.6 s^{-1}

$\tau_0 = 5.6\cdot10^{-16}\pm 3.6\cdot10^{-16}$ s

Δ_{eff}/k_B = 149 K (fixed)

R^2 = 0.80783



Orbach + Raman+QTM

$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B+C_{Raman}T^{nRaman}$

B = 119.7±1.5 s^{-1}

C_{Raman} = 5.42±0.32 $s^{-1}K^{-nRaman}$

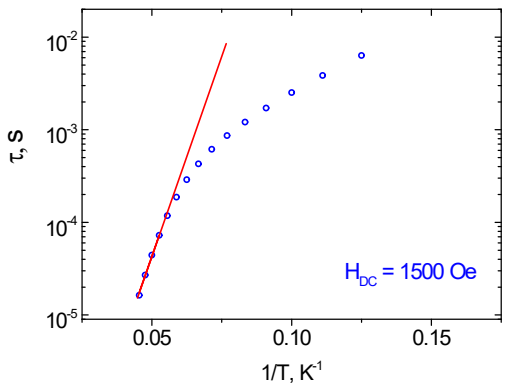
n_{Raman} = 2.5±0.03

$\tau_0 = 4.1\cdot10^{-8} \pm 1.4\cdot10^{-8}$ s

Δ_{eff}/k_B = 148.8 K (fixed)

R^2 = 0.99975

H_{DC} = 1500 Oe



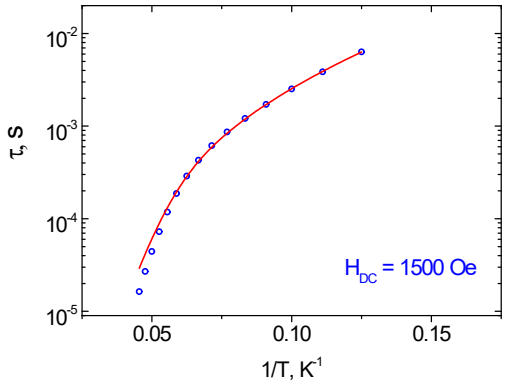
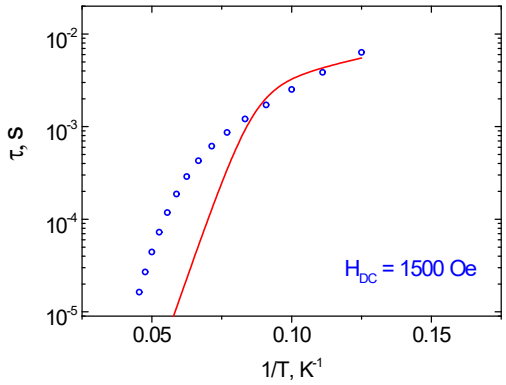
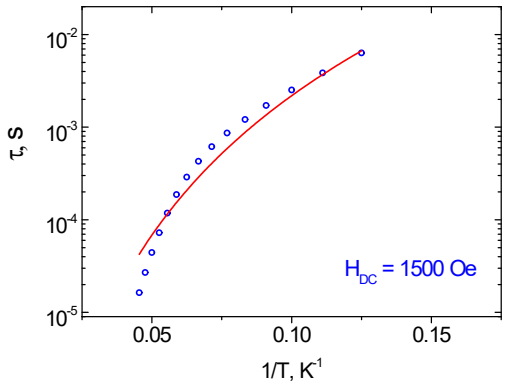
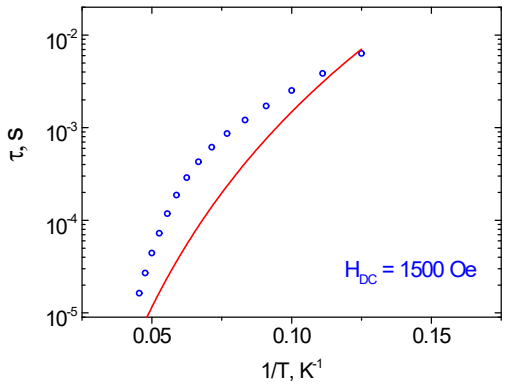
Orbach

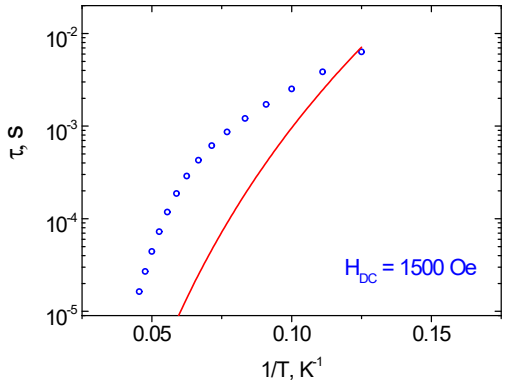
$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)$

$\tau_0 = 2.06\cdot10^{-9}\pm 6.8\cdot10^{-10}$ s

Δ_{eff}/k_B = 199 ± 6 K

R^2 = 0.99773

	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 0.033 \pm 0.002\ s^{-1}K^{-nRaman}$ $n_{Raman} = 4.07 \pm 0.03$ $\tau_0 = 4.8 \cdot 10^{-9} \pm 8 \cdot 10^{-10}$ s $\Delta_{eff}/k_B = 199$ K (fixed) $R^2 = 0.99984$
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 2.8 \pm 0.2\ s^{-1}K^{-nRaman}$ $n_{Raman} = 2$(fixed) $\tau_0 = 9 \cdot 10^{-11} \pm 4 \cdot 10^{-11}$ s $\Delta_{eff}/k_B = 199$ K (fixed) $R^2 = 0.94342$
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 4.6 \cdot 10^{-3} \pm 1 \cdot 10^{-4}\ s^{-1}K^{-nRaman}$ $n_{Raman} = 5$ (fixed) $\tau_0 = 1 \pm 3.8 \cdot 10^{8}$ s $\Delta_{eff}/k_B = 199$ K (fixed) $R^2 = 0.98443$
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 6.8 \cdot 10^{-5} \pm 5 \cdot 10^{-6}\ s^{-1}K^{-nRaman}$ $n_{Raman} = 7$ (fixed) $\tau_0 = 1 \pm 1.7 \cdot 10^{10}$ s $\Delta_{eff}/k_B = 199$ K (fixed) $R^2 = 0.88967$



Raman+Orbach

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

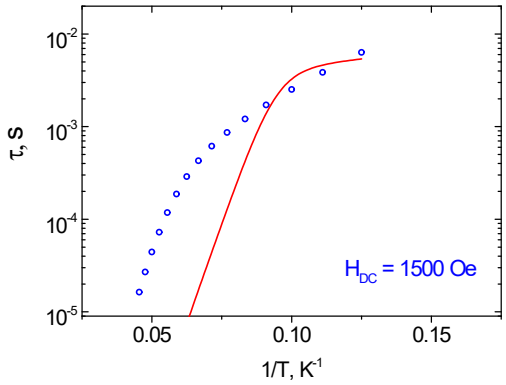
$C_{Raman} = 1.0 \cdot 10^{-6} \pm 1 \cdot 10^{-7}\ s^{-1} K^{-nRaman}$

$n_{Raman} = 9$ (fixed)

$\tau_0 = 1 \pm 1.8 \cdot 10^{11}$ s

$\Delta_{eff}/k_B = 199$ K (fixed)

$R^2 = 0.78341$



Direct+Orbach

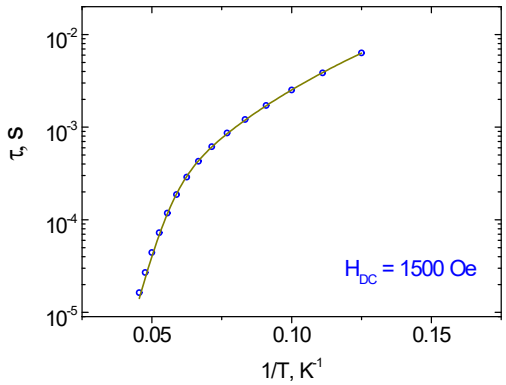
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + A_{direct} T H^4$

$A_{direct} = 4.6 \cdot 10^{-12} \pm 3.62 \cdot 10^{-13}\ K^{-1} Oe^{-4} s^{-1}$

$\tau_0 = 3 \cdot 10^{-11} \pm 1 \cdot 10^{-11}$ s

$\Delta_{eff}/k_B = 199$ K (fixed)

$R^2 = 0.90608$



Raman+Orbach

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

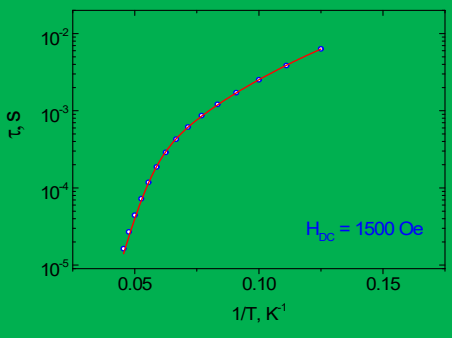
$C_{Raman} = 0.031 \pm 0.0013\ s^{-1} K^{-nRaman}$

$n_{Raman} = 4.1 \pm 0.019$

$\tau_0 = 9.6 \cdot 10^{-11} \pm 1.6 \cdot 10^{-10}$ s

$\Delta_{eff}/k_B = 265 \pm 28.7$ K

$R^2 = 0.99993$



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

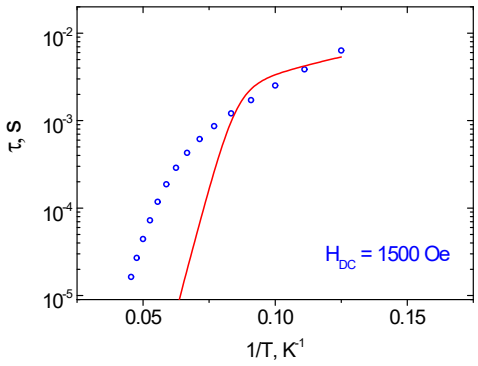
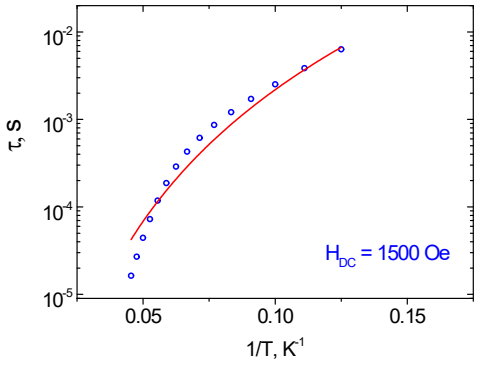
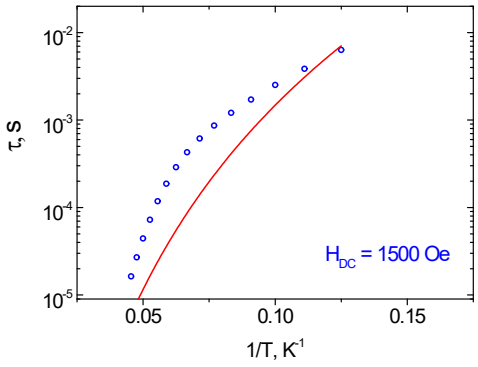
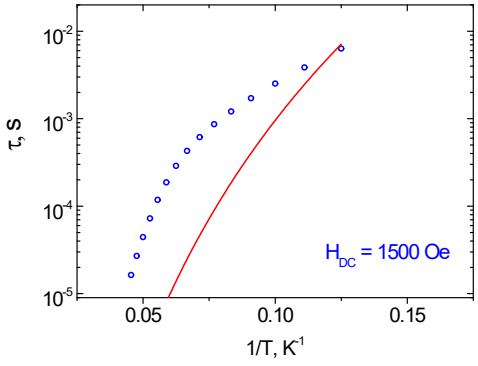
$C_{raman} = 0.03 \pm 0.001 s^{-1} K^{-nRaman}$

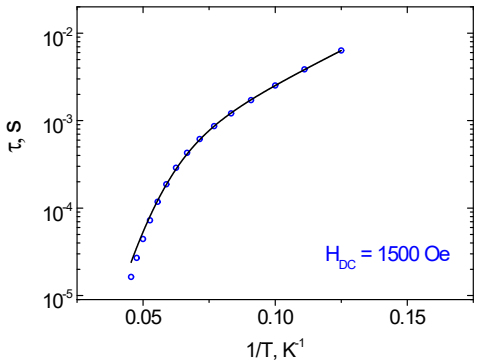
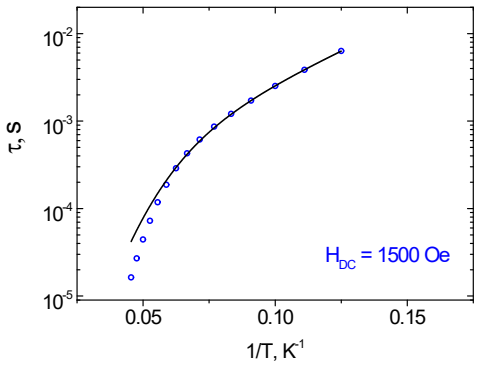
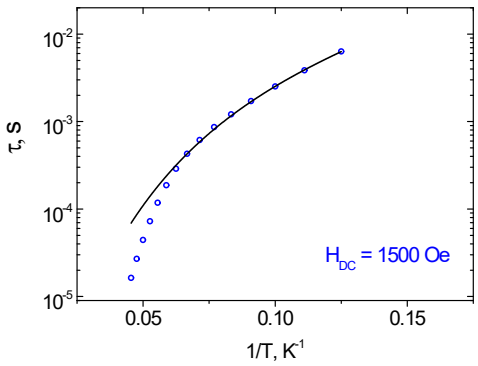
$n_{raman} = 4.1 \pm 0.016$

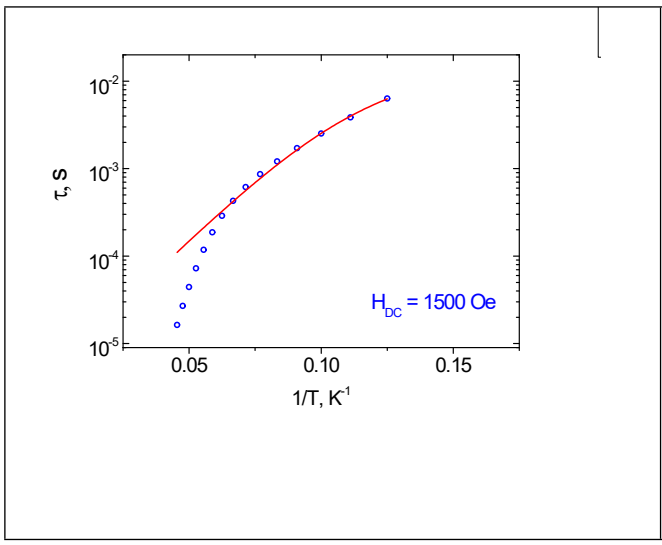
$\Delta_{eff}/k_B = 265$ K (fixed, from ab initio results)

$\tau_0 = 9.5 \cdot 10^{-11} \pm 1.2 \cdot 10^{-11}$ s

$R^2 = 0.99993$

	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = 2.9·± 0.2s^{-1}K$^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 4.2 \cdot 10^{-13} \pm 2.8 \cdot 10^{-13}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.92499
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = 4.6·= $^{-3}$·± 1·10^{-4} s^{-1}K$^{-nRaman}$ n_{Raman} = 5 (fixed) $\tau_0 = 1 \pm 1.16 \cdot 10^{10}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.98443
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = 6.8·10^{-5}·± 5·10^{-6} s^{-1}K$^{-nRaman}$ n_{Raman} = 7 (fixed) $\tau_0 = 1 \pm 9.5 \cdot 10^{11}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.88967
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = 1.0·10^{-6}·± 1·10^{-7} s^{-1}K$^{-nRaman}$ n_{Raman} = 9 (fixed) $\tau_0 = 1 \pm 2.1 \cdot 10^{13}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.78342

	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 3.3 \cdot 10^{-8} \pm 1 \cdot 10^{-9} s^{-1} K^{-nRaman}$ $n_{Raman} = 9$ (fixed) $\tau_0 = 8.6 \cdot 10^{-5} \pm 2 \cdot 10^{-6}$ s $\Delta_{eff}/k_B = 34.6 \pm 0.19$ K **$R^2 = 0.99996$**
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 8.9 \cdot 10^{-6} \pm 5.1 \cdot 10^{-7} s^{-1} K^{-nRaman}$ $n_{Raman} = 7$ (fixed) $\tau_0 = 1.4 \cdot 10^{-4} \pm 1.1 \cdot 10^{-5}$ s $\Delta_{eff}/k_B = 31.4 \pm 0.6$ K **$R^2 = 0.99981$**
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 0.00273 \pm 2.4 \cdot 10^{-4}$ $s^{-1} K^{-nRaman}$ $n_{Raman} = 5$ (fixed) $\tau_0 = 8.02 \cdot 10^{-4} \pm 5 \cdot 10^{-4}$ s $\Delta_{eff}/k_B = 23.1 \pm 4.2$ K **$R^2 = 0.99918$**



Raman+Orbach

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

C_{Raman} = 1.9±0.1 $s^{-1}K^{-nRaman}$

n_{Raman} = 2 (fixed)

$\tau_0 = 5.7 \cdot 10^{-6} \pm 2.1 \cdot 10^{-6}$ s

Δ_{eff}/k_B = 67.4±4.4 K

R^2 = 0.99724

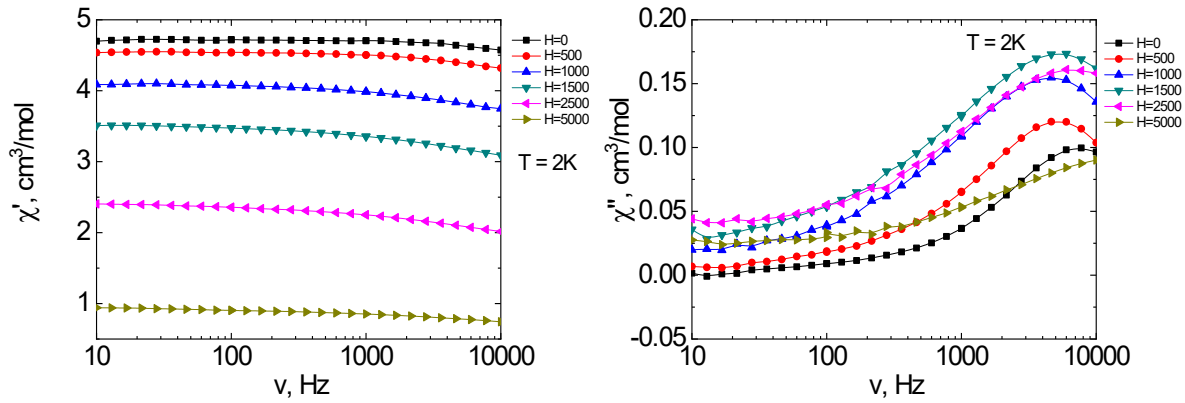


Fig. S12. Frequency dependences of real (left) and imaginary (right) components of dynamic magnetic susceptibility of complex **2** at 2 K in various DC magnetic fields. Solid lines are visual guides.

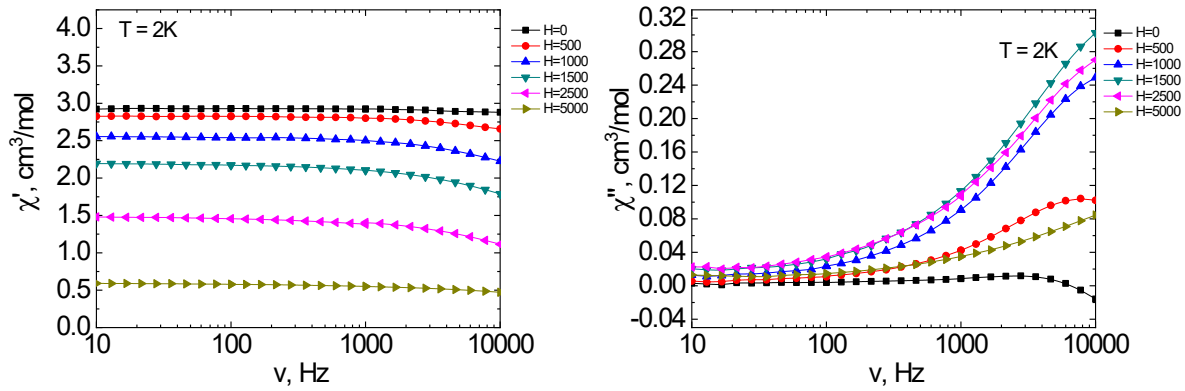


Fig. S13. Frequency dependences of real (left) and imaginary (right) components of magnetic susceptibility of complex **4** at 2 K in various DC magnetic fields. Solid lines are visual guides.

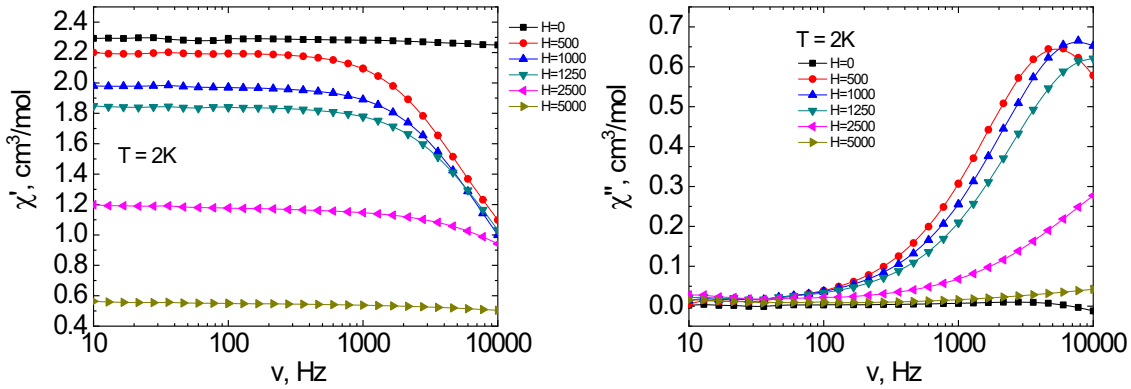


Fig. S14. Frequency dependencies of real (left) and imaginary (right) components of dynamic magnetic susceptibility of complex **3** at 2 K in various DC magnetic fields. Solid lines are visual guides.

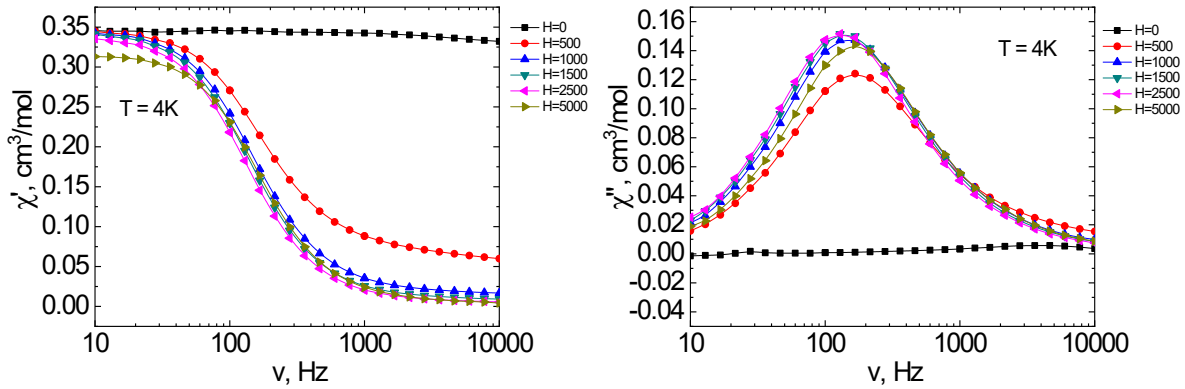


Fig. S15. Frequency dependencies of real (left) and imaginary (right) components of dynamic magnetic susceptibility of complex **5** at 4 K in various DC magnetic fields. Solid lines are visual guides.

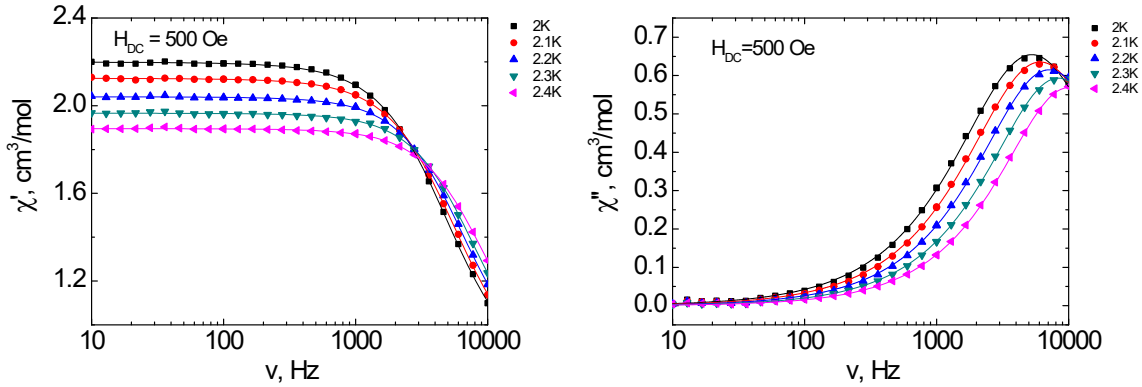


Fig. S16. Frequency dependencies of real (left) and imaginary (right) components of magnetic susceptibility for complex **3** in 500 Oe DC magnetic field. Solid lines represent best fit by the generalized Debye model.

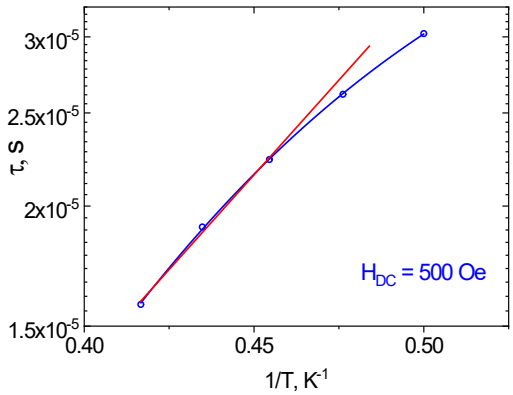


Fig. S17. Plots of the τ(1/T) dependency for complex **3** in 500 Oe. Red line represents the best fit by Orbach mechanism while blue line represents the best fit by the sum of Orbach and direct relaxation mechanisms.

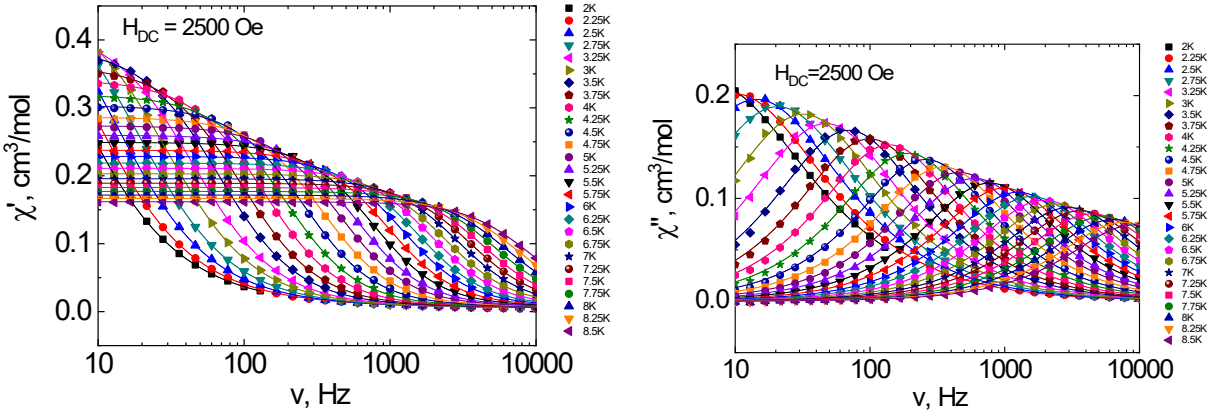


Fig. S18. Frequency dependencies of real (left) and imaginary (right) components of magnetic susceptibility for complex **5** in 2500 Oe DC magnetic field. Solid lines represent best fit by the generalized Debye model.

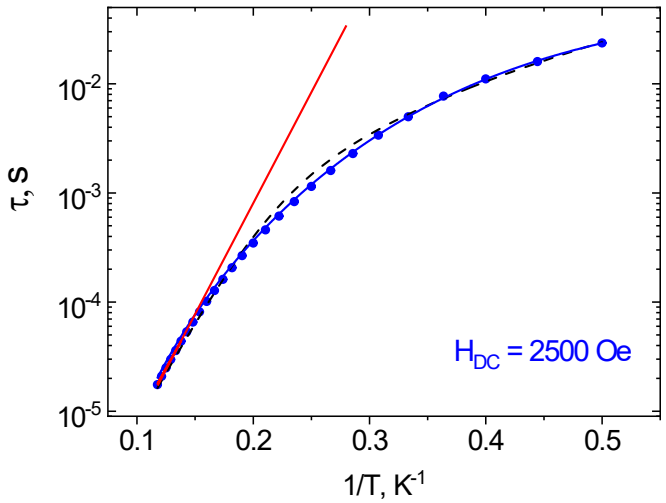


Fig. S19. Plots of the τ(1/T) dependency for complex **5** in 2500 Oe. Red, dashed black and blue lines respectively represent best fit by Orbach, Raman+Orbach and direct+Raman relaxation mechanisms.

Table S6. CASSCF computed energy levels (K, Fig. 4a), the composition of the effective g′-tensor (assuming a pseudospin S = ½)and major (>10%) components of the wave function for each m_J state of the lowest atomic multiplet J=15/2 of Dy^{3+} in complex **1**. KDs involved in magnetic relaxation are specified.

KD	E, K	g_x	g_y	g_z	Wave function*, %
1	0.0	0.02	0.04	18.92	84|±15/2>+12|±11/2>
2	150.7	0.23	0.37	14.89	63|±13/2>+29|±9/2>
3	265.9	0.15	0.66	11.51	40|±7/2>+36|±11/2>
4	354.7	7.61	6.31	4.75	42|±5/2>+20|±1/2>+13|±13/2>+11|±9/2>
5	436.6	2.01	3.03	11.59	40|±3/2>+16|±1/2>+14|±11/2>+11|±9/2>
6	570.5	0.19	0.22	15.66	41|±1/2>+16|±9/2>+15|±7/2>+11|±3/2>
7	720.9	0.02	0.05	18.98	29|±5/2>+26|±3/2>+19|±7/2>+15|±1/2>
8	810.3	0.01	0.01	19.70	22|±11/2>+22|±9/2>+17|±7/2>+13|±13/2>+10|±5/2>

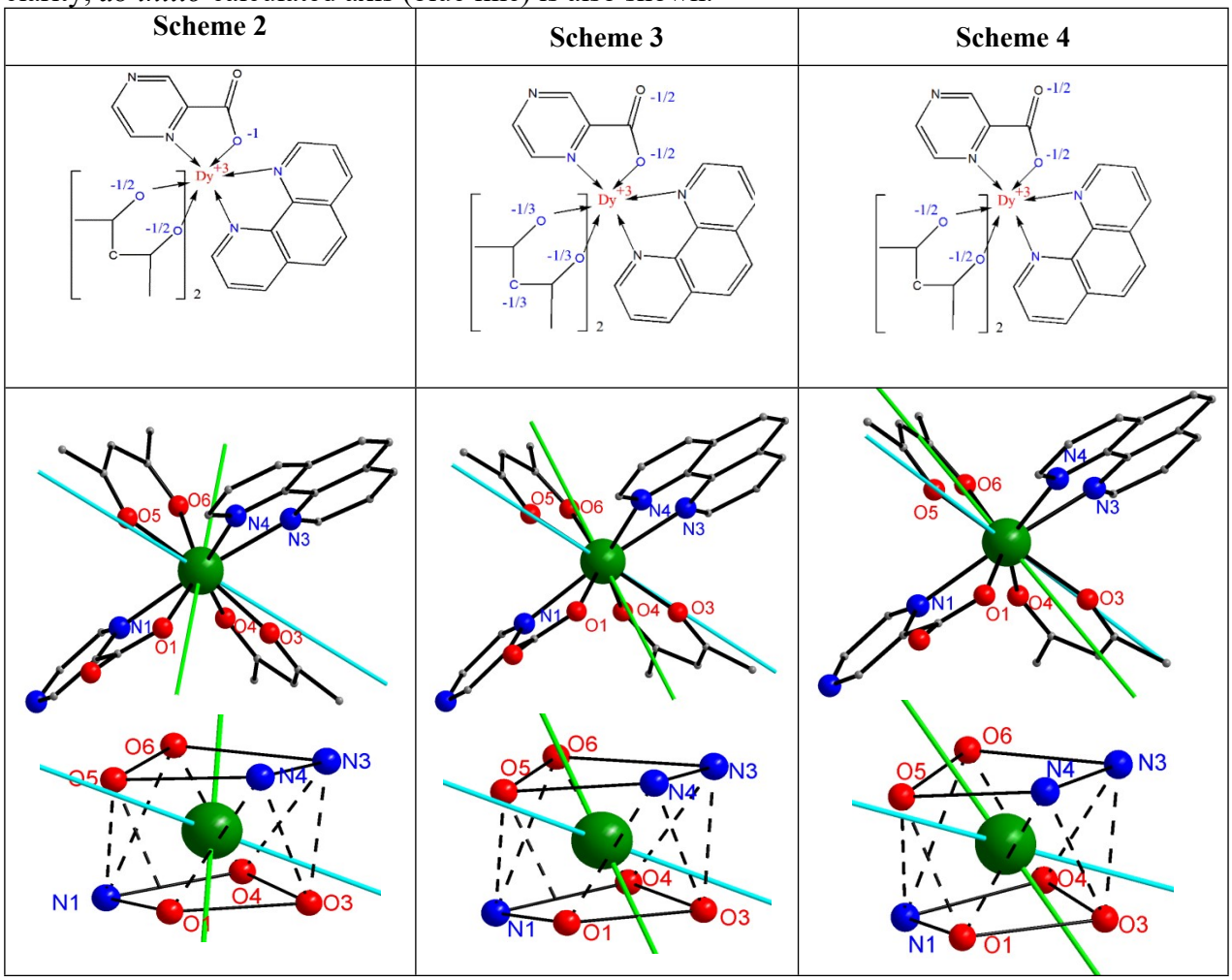
Table S7. CASSCF computed energy levels (K, Fig. 4b), the composition of the effective g′-tensor (assuming a pseudospin S = ½) and major(>10%) components of the wave function for each m_J state of the lowest atomic multiplet J=15/2 of Er^{3+} in complex **3**. KDs involved in magnetic relaxation are specified.

KD	Energy, K	g_x	g_y	g_z	Wave function*, %
1	0.0	1.02	1.61	14.50	37|±13/2>+28|±15/2>+24|±11/2>
2	28.9	1.53	2.40	12.97	36|±13/2>+34|±11/2>+10|±15/2>
3	104.7	2.94	3.94	8.23	40|±9/2>+20|±3/2>+11|±1/2>
4	133.4	0.73	1.38	13.12	33|±7/2>+21|±5/2>+16|±15/2>+10|±9/2>
5	208.8	1.24	4.53	8.85	29|±15/2>+17|±7/2>+12|±5/2>+11|±11/2>+10|±9/2>
6	261.8	2.05	2.25	9.22	21|±5/2>+18|±3/2>+16|±9/2>+14|±1/2>+12|±11/2>
7	360.5	1.70	2.18	11.83	35|±1/2>+20|±7/2>+15|±3/2>+12|±5/2>
8	449.6	0.38	0.94	15.65	34|±3/2>+26|±5/2>+24|±1/2>+10|±7/2>

Table S8. CASSCF computed energy levels (K, Fig. 4c), the composition of the effective g′-tensor (assuming a pseudospin S = ½) and major (>10%) components of the wave function for each m_J state of the lowest atomic multiplet J=7/2 of Yb^{3+} in complex **5**. KDs involved in magnetic relaxation are specified.

KD	Energy, K	g_x	g_y	g_z	Wave function*, %
1	0.0	0.05	0.19	7.52	92|±7/2>
2	490.0	0.76	1.00	5.16	92|±5/2>
3	640.0	0.64	1.70	6.63	67|±3/2>+18|±1/2>
4	747.8	0.64	1.70	6.63	72|±1/2>+25|±3/2>

Fig. S20. Alternative schemes of partial charges distribution assigned to complex **1** for MAGELLAN calculations and corresponding positions of the anisotropy axis (green line). For clarity, *ab initio* calculated axis (blue line) is also shown.



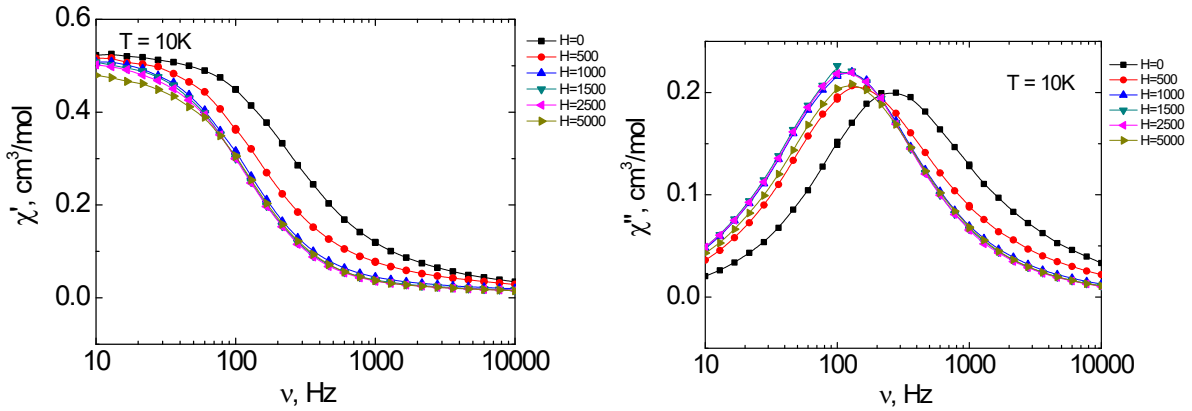


Fig. S21. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ", right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.5}$** in various DC magnetic fields at 10 K. Solid lines are visual guides.

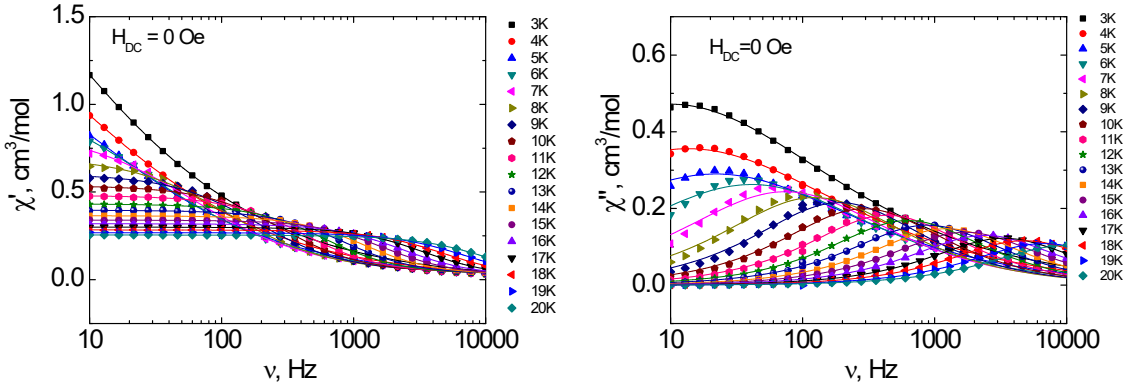


Fig. S22. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ", right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.5}$** in zero DC magnetic field. Solid lines shows approximations of experimental data for one relaxation mode by the generalized Debye model.

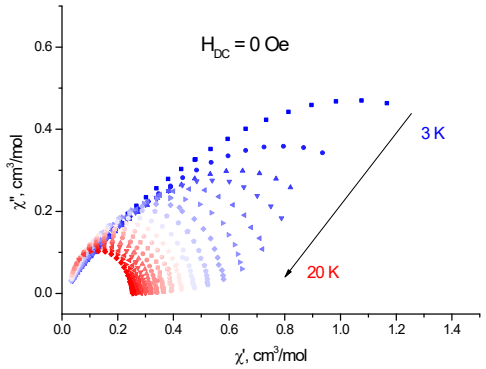


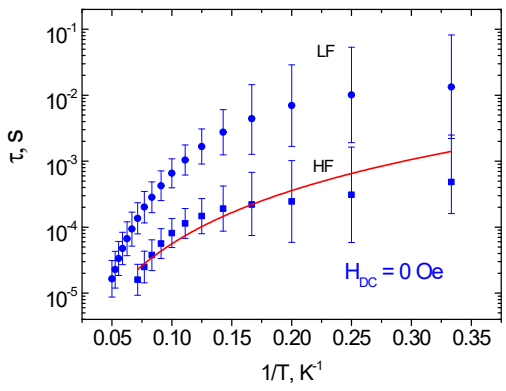
Fig. S23. Cole-Cole plots for **1_Y$_{0.5}$** in zero DC field.

Table. S9. Distribution of parameter α at H_{DC} = 0 Oe for **1_Y$_{0.5}$**.

T, K	1 Relax	2 Relax
3	0.50121	0.38295
4	0.48555	0.35196
5	0.44752	0.30003
6	0.37238	0.23593
7	0.28503	0.13922

8	0.217	0.09296
9	0.17226	0.07141
10	0.1477	0.06703
11	0.12685	0.07037
12	0.115	0.07423
13	0.11291	0.07827
14	0.09588	0.07564
15	0.09531	0.08877
16	0.08826	0.08771
17	0.0845	0.08186
18	0.08718	0.08996
19	0.0899	0.09359
20	0.10356	0.10001

Table S10. Fitting of the τ *vs. 1/T* dependence for the HF process in **1_Y$_{0.5}$** (H_{DC} = 0 Oe, T = 3-20 K). Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color.



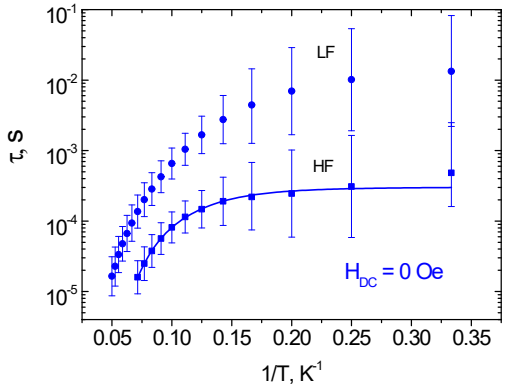
Raman

$\tau^{-1} = C_{Raman} T^{nRaman}$

C_{Raman} = 37.6 ± 26 $s^{-1}K^{-nRaman}$

n_{Raman} = 2.7 ± 0.3

R^2 = 0.72064



QTM+Raman+Orbach

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + B + C_{Raman} T^{nRaman}$

B = 3292 ± 337 s^{-1}

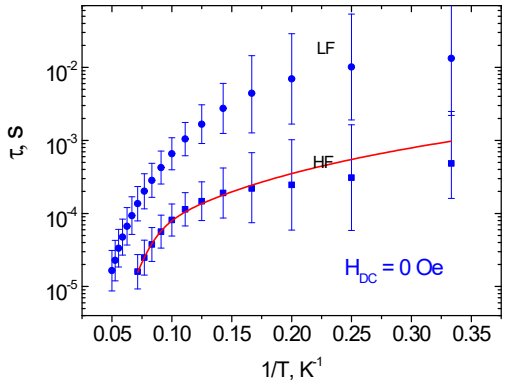
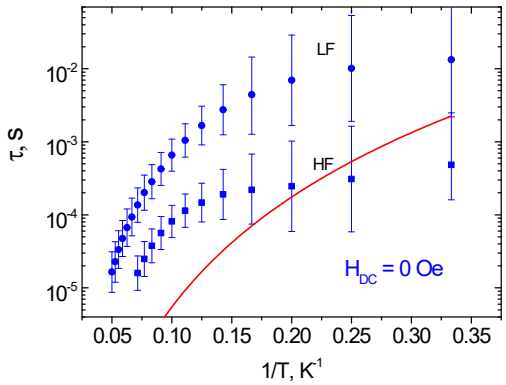
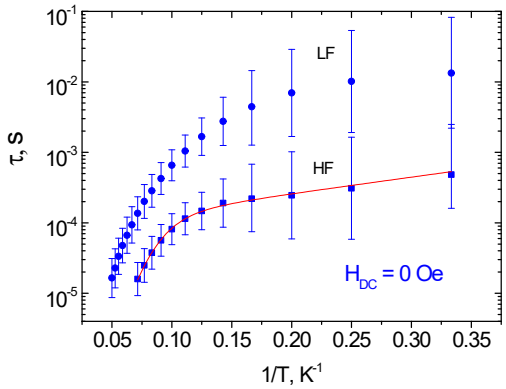
C_{Raman} = 0.5 ± 0.46 $s^{-1}K^{-nRaman}$

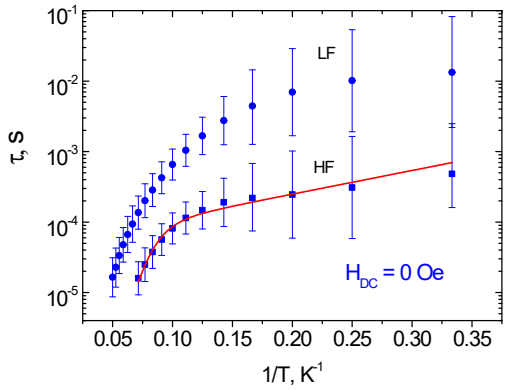
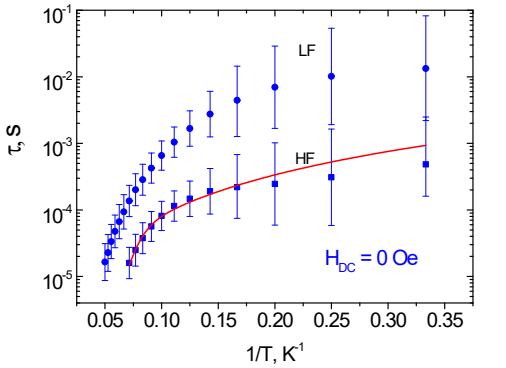
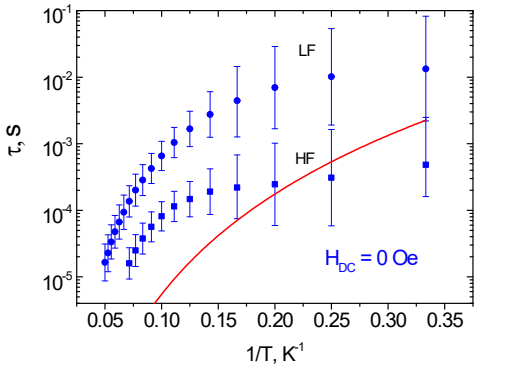
n_{Raman} = 4.24 ± 0.39

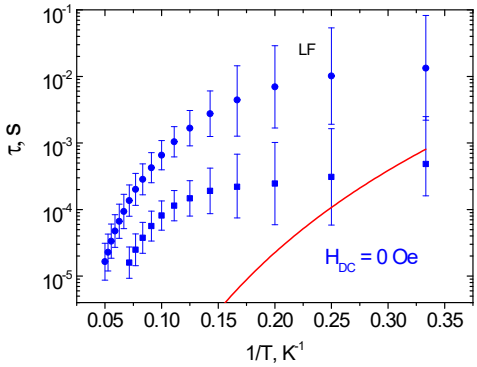
$\tau_0 = 1.02 \cdot 10^{-9} \pm 2.31 \cdot 10^{-10}$ s

Δ_{eff}/k_B = 149 K (fixed)

R^2 = 0.99709

	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman}$ C_{Raman} = *9.45 ± 0.83* $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) τ_0 = $2 \cdot 10^{-7}$ ± $5.3 \cdot 10^{-8}$ s Δ_{eff}/k_B = 93.6 ± 4.3 K R^2 = 0.97857
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman}$ C_{Raman} = *1.83 ± 1.7* $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) τ_0 = $1 \cdot 10^{-7}$ ± $4.4 \cdot 10^{11}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = -1.54898
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman}$ C_{Raman} = $5.35 \cdot 10^{-4}$ ± $1.13 \cdot 10^{-5}$ $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) τ_0 = $5.35 \cdot 10^{-4}$ ± $1.13 \cdot 10^{-5}$ s Δ_{eff}/k_B = 5.38 ± 0.5 K R^2 = 0.99767

	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 2.9 \cdot 10^{-6} \pm 2.3 \cdot 10^{-7}$ $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) $\tau_0 = 5.3 \cdot 10^{-5} \pm 8.9 \cdot 10^{-6}$ s Δ_{eff}/k_B = 7.72 ± 1.2 K R^2 = 0.97431
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 119.2 \pm 5.6$ $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 5.43 \cdot 10^{-10} \pm 6.65 \cdot 10^{-11}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.96789
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 1.83 \pm 1.68$ $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) $\tau_0 = 2.8 \cdot 10^{8}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = -1.54898



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman}$

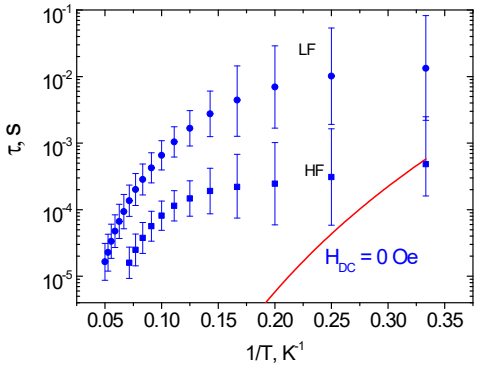
C_{Raman} = *0.57 ± 1.68* $s^{-1}K^{-nRaman}$

n_{Raman} = 7 (fixed)

$\tau_0 = 2.2 \cdot 10^7$ s

Δ_{eff}/k_B = 149 K (fixed)

R^2 = -1.82232



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman}$

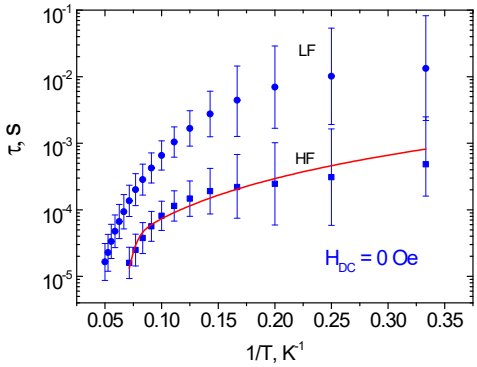
C_{Raman} = *0.08831 ± 0.37327* $s^{-1}K^{-nRaman}$

n_{Raman} = 9 (fixed)

τ_0 = 1 s

Δ_{eff}/k_B = 149 K (fixed)

R^2 = -1.83881



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman}$

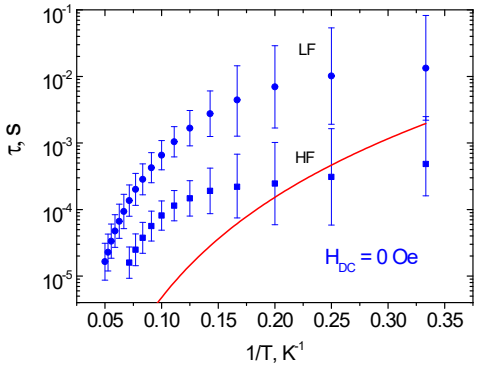
C_{Raman} = *136.03 ± 9.59* $s^{-1}K^{-nRaman}$

n_{Raman} = 2 (fixed)

$\tau_0 = 1.23 \cdot 10^{-13} \pm 3.12 \cdot 10^{-14}$ s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = 0.90895



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

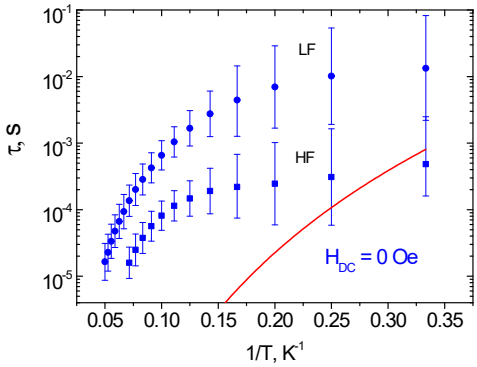
C_{Raman} = 2.1 ± 2.2 $s^{-1}K^{-nRaman}$

n_{Raman} = 5 (fixed)

τ_0 = 0.056 ± 1.02·10^{13} s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = -1.55409



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

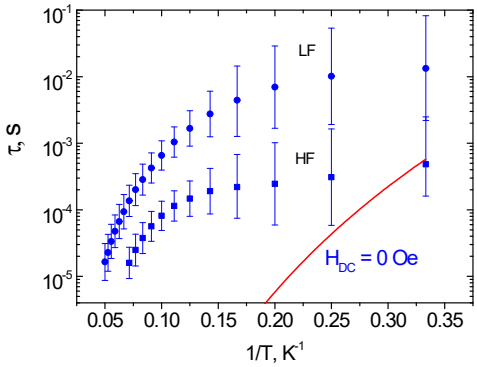
C_{Raman} = 0.56768 ± 1.68033 $s^{-1}K^{-nRaman}$

n_{Raman} = 7 (fixed)

τ_0 = 1 s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = -1.82232



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

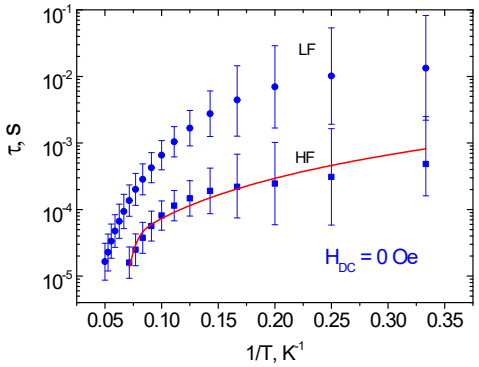
C_{Raman} = 0.08831 ± 0.37327 $s^{-1}K^{-nRaman}$

n_{Raman} = 9 (fixed)

τ_0 = 1 s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = -1.83881



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

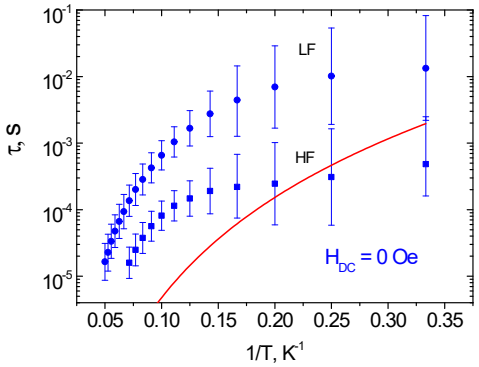
C_{Raman} = *136.03 ± 9.59* $s^{-1}K^{-nRaman}$

n_{Raman} = 2 (fixed)

$\tau_0 = 1.23 \cdot 10^{-13} \pm 3.12 \cdot 10^{-14}$ s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = 0.90895



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

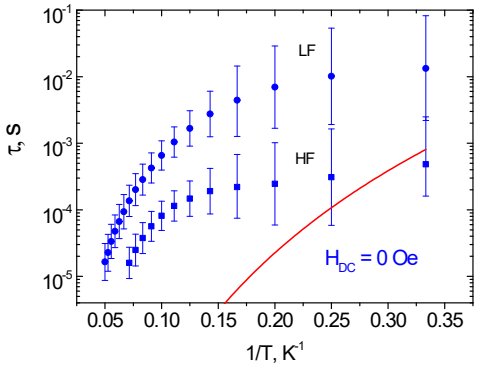
C_{Raman} = *2.1 ± 2.2* $s^{-1}K^{-nRaman}$

n_{Raman} = 5 (fixed)

$\tau_0 = 0.056 \pm 1.02 \cdot 10^{13}$ s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = -1.55409



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$

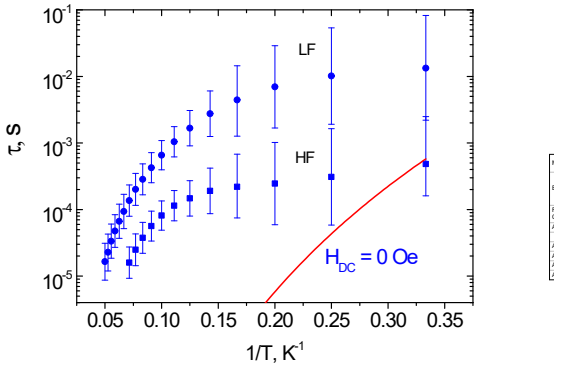
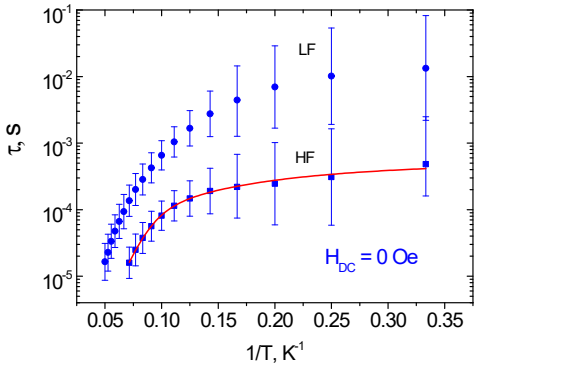
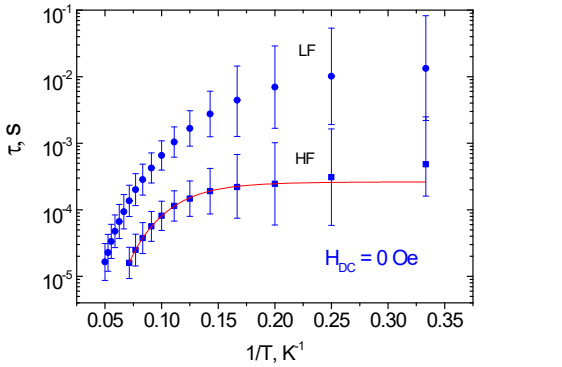
C_{Raman} = *0.56768 ± 1.68033* $s^{-1}K^{-nRaman}$

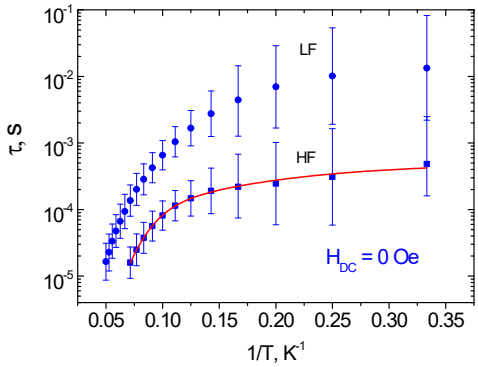
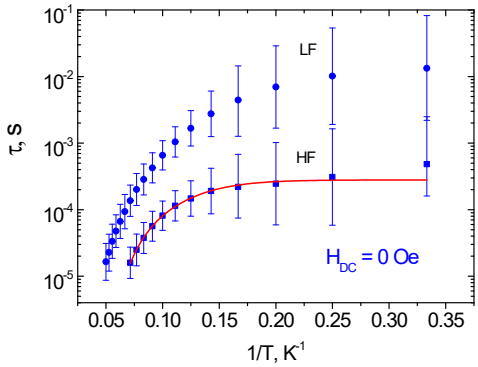
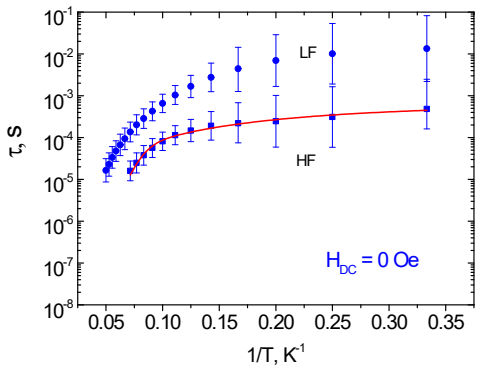
n_{Raman} = 7 (fixed)

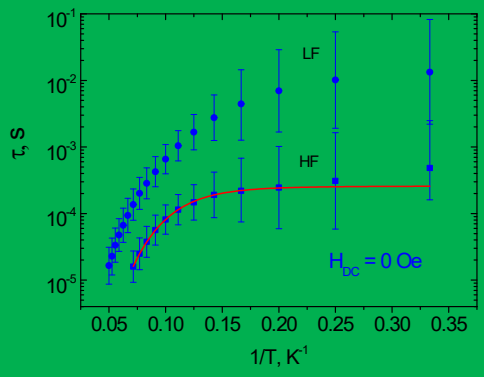
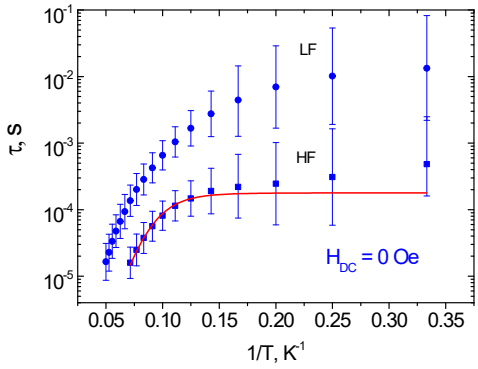
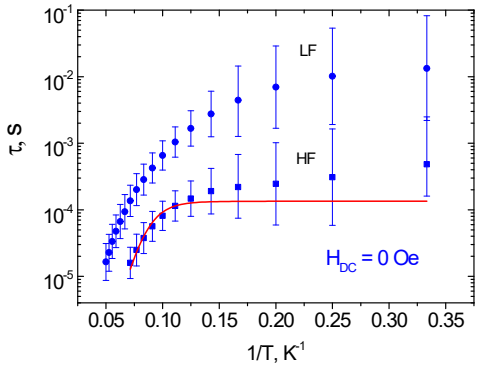
τ_0 = 1 s

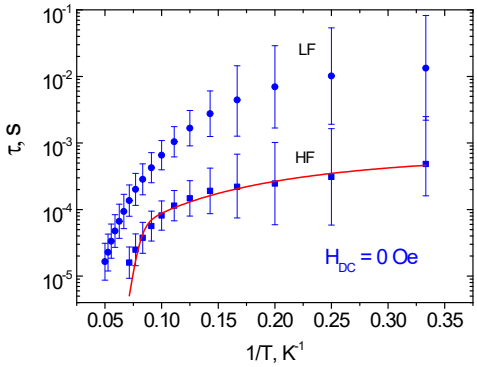
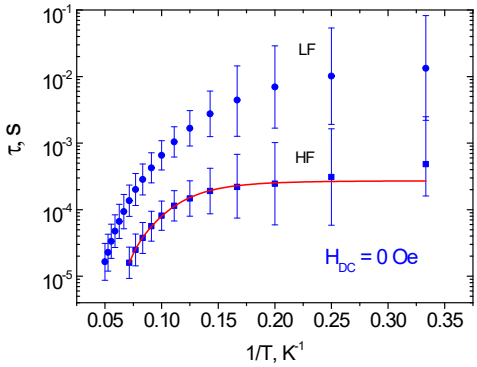
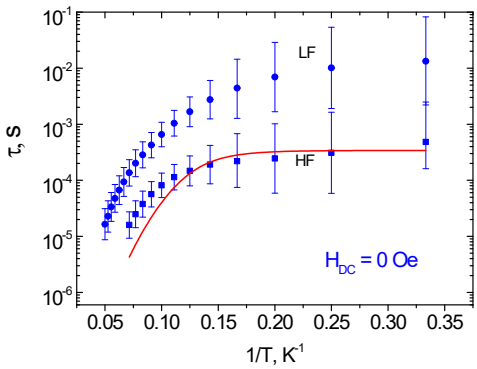
Δ_{eff}/k_B = 265 K (fixed)

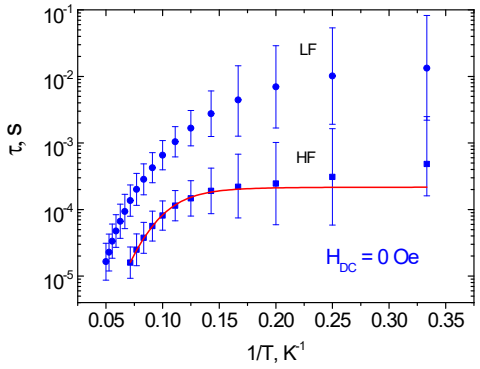
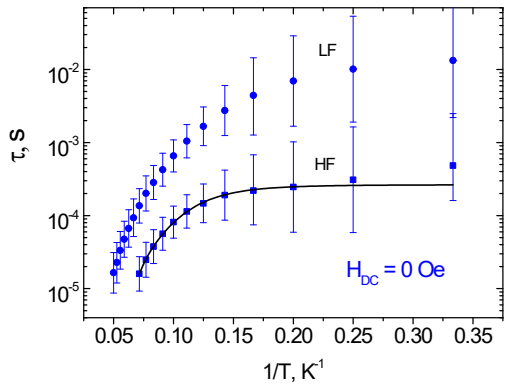
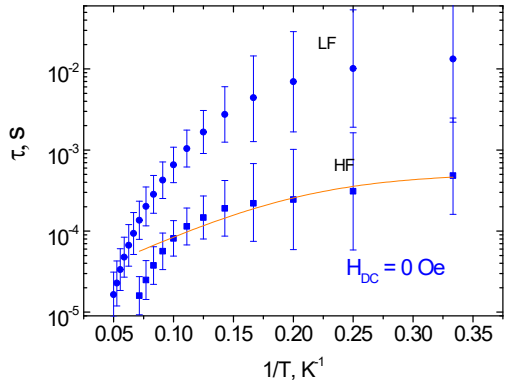
R^2 = -1.82232

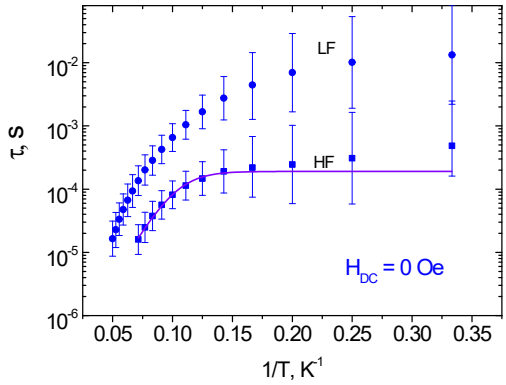
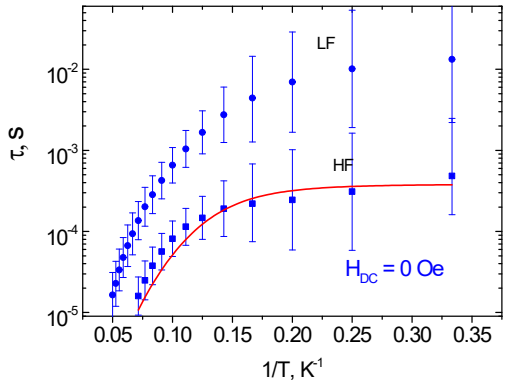
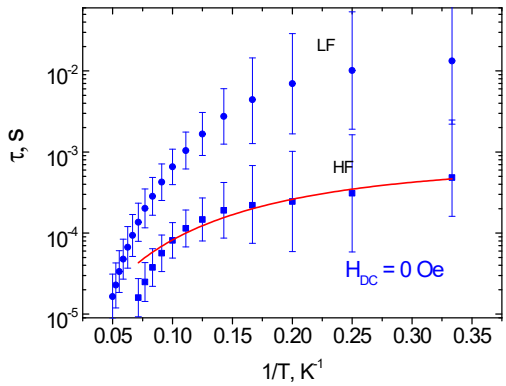
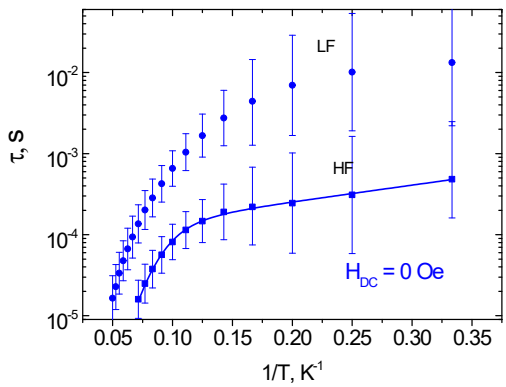
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = *0.08831 ± 0.37327* $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) τ_0 = 1 s Δ_{eff}/k_B = 265 K (fixed) R^2 = -1.83881
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}+B$ B =*1740 ± 236* s^{-1} C_{Raman} = *75.3 ± 4.9* $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 2.17\cdot10^{-8} \pm 6.0\cdot10^{-9}$ s Δ_{eff}/k_B = 97 ± 3.8 K R^2 = 0.99842
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}+B$ B = 3791 *± 213* s^{-1} C_{Raman} = *0.085 ± 0.0047* $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) $\tau_0 = 1.45\cdot10^{-10} \pm 6.86\cdot10^{-10}$ s Δ_{eff}/k_B = 184 ± 68.2 K R^2 = 0.99623

	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$ B = 2058 ± 424 s^{-1} C_{Raman} = 5.12·10^{-4} ± 1.2·10^{-5} $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) τ_0 = 6.24·10^{-5} ± 1.24·10^{-5} s Δ_{eff}/k_B = 11.7 ± 2.3 K R^2 = 0.99862
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$ B = 3575 ± 362 s^{-1} C_{Raman} = 2·10^{-6}± 1.9·10^{-7} $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) τ_0 = 4.6·10^{-6} ± 2.1·10^{-6} s Δ_{eff}/k_B = 34.6 ± 4.6 K R^2 = 0.99631
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$ B = 1390 ± 167 s^{-1} C_{Raman} = 93.58 ± 7.73 $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) τ_0 = 4.06·10^{-10} ± 1.2·10^{-10} s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.98379

	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman} + B$ B = 3875 ± 187 s^{-1} C_{Raman} = 0.08229 ± 0.00329 $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) $\tau_0 = 1.7\cdot10^{-9} \pm 3.3\cdot10^{-10}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.99642
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman} + B$ B = 5599 ± 476 s^{-1} $C_{Raman} = 5.87\cdot10^{-4} \pm 7.8\cdot10^{-5}s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) $\tau_0 = 1 \pm 4.5\cdot10^{8}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.97899
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman} + B$ B = 7437 ± 1093 s^{-1} $C_{Raman} = 3.4\cdot10^{-6} \pm 1.8\cdot10^{-6}s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) $\tau_0 = 1 \pm 1.8\cdot10^{9}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.91981

	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$ B = 1260 ± 150 s^{-1} C_{Raman} = 100.4 ± 9.4 $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 3.4 \cdot 10^{-14} \pm 3.9 \cdot 10^{-14}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.97539
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$ B = 3681 ± 170 s^{-1} C_{Raman} = 0.0885 ± 0.00246 $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) $\tau_0 = 5.02 \cdot 10^{-13} \pm 1.05 \cdot 10^{-13}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.99616
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$ B = 2928 ± 326 s^{-1} C_{Raman} = 0.0022 ± 0.00106 $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) $\tau_0 = 1 \pm 3.8 \cdot 10^{14}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.78521

	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + +C_{Raman}T^{nRaman}+B$ B = 4618 ± 231 s^{-1} C_{Raman} = 0.00622 ± 2.4·10^{-4} $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) τ_0 = 0.99753 ± 6.75·10^{11} s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.99266
	**QTM+Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + +B+C_{Raman}T^{nRaman}$ B = 3784.9 ± 322 s^{-1} *C_{Raman} = 0.067 ± 0.056* $s^{-1}K^{-nRaman}$ *n_{Raman} = 5.111 ± 0.35* τ_0 = 5.6·10^{-13} ±2.41·10^{-13} s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.99575
	**QTM+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B$ B = 1935.6±264.6 s^{-1} τ_0 = 1.98·10^{-5} ± 8.9·10^{-6} s Δ_{eff}/k_B = 16.2 ± 3.2 K R^2 = 0.93563

	**QTM+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + B$ B = 5243 ± 424 s^{-1} $\tau_0 = 1.13 \cdot 10^{-7} \pm 3.9 \cdot 10^{-8}$ s Δ_{eff}/k_B = 71.4 ± 4.3 K R^2 = 0.9849
	**QTM+Raman** $\tau^{-1} = C_{Raman} T^{nRaman} + B$ *B = 2614 ± 243 s^{-1}* *C_{Raman} = 0.17 ± 0.05 $s^{-1}K^{-nRaman}$* *n_{Raman} = 5 (fixed)* *R^2 = 0.87484*
	**QTM+Raman (parameters refined)** $\tau^{-1} = C_{Raman} T^{nRaman} + B$ *B = 1260 ± 471 s^{-1}* *C_{Raman} = 89.8 ± 82.4 $s^{-1}K^{-nRaman}$* *n_{Raman} = 2.08 ± 0.4* *R^2 = 0.96282*
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $C_{Raman} = 5.58 \cdot 10^{-4} \pm 5.94 \cdot 10^{-6}$ $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) $\tau_0 = 9.92 \cdot 10^{-5} \pm 4.43 \cdot 10^{-5}$ s Δ_{eff}/k_B = 4.7 ±0.16 K R^2 = 0.9972

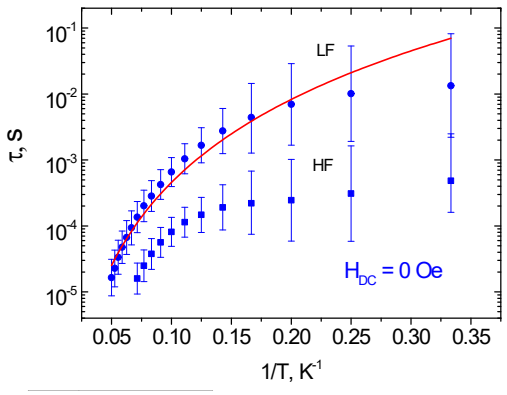
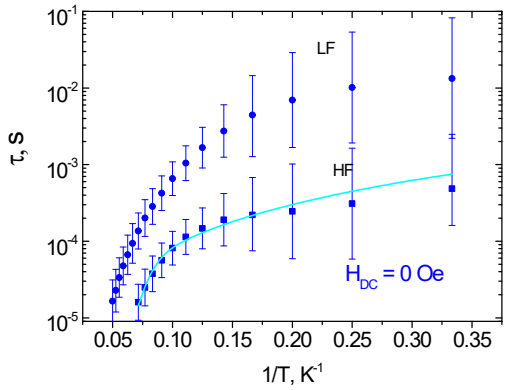
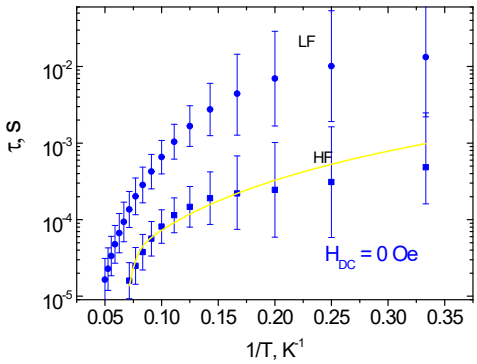
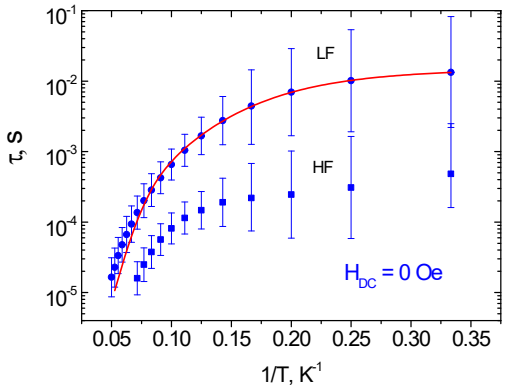
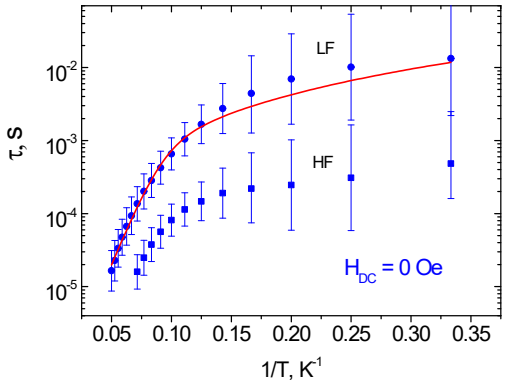
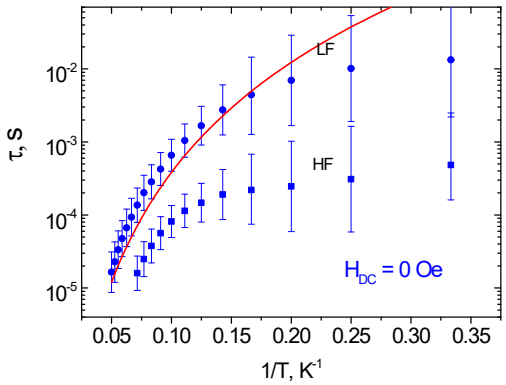
	**Raman** $\tau^{-1} = C_{Raman}T^{nRaman}$ C_{Raman} = 0.15 ± 0.07 $s^{-1}K^{-nRaman}$ n_{Raman} = 4.18 ± 0.19 R^2 = 0.82815
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = 186.2 ± 76.2 $s^{-1}K^{-nRaman}$ n_{Raman} = 1.79 ± 0.19 $\tau_0 = 4.93\cdot10^{-10} \pm 6.47\cdot10^{-11}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.96992
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = 94.3 ± 53.6 $s^{-1}K^{-nRaman}$ $n_{Raman} = 2.16\pm0.25$ $\tau_0 = 1.3\cdot10^{-13} \pm 4.1\cdot10^{-14}$ s Δ_{eff}/k_B = 265 K(fixed) R^2 = 0.90394

Table S11. Fitting of the *τ vs. 1/T* dependence for the LF process in **1_Y$_{0.5}$**(H_{DC} = 0 Oe, T = 3-20 K). Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color.

	**QTM+Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B+C_{Raman}T^{nRaman}$ B = 65 ± 0.6 s^{-1} C_{Raman} = *0.13 ± 0.014* $s^{-1}K^{-nRaman}$ n_{Raman} = *3.98 ± 0.06* $\tau_0 = 5.07\cdot10^{-9} \pm 2.18\cdot10^{-9}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.99982
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = *113.9 ± 7.2* $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 3.53\cdot10^{-9} \pm 5.7\cdot10^{-9}$ s Δ_{eff}/k_B = 123.7 ± 21.5 K R^2 = 0.96975
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ **LF** C_{Raman} = *0.026 ± 0.005* $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) τ_0 = 0.0421 s Δ_{eff}/k_B = 3000 K R^2 = 0.32713

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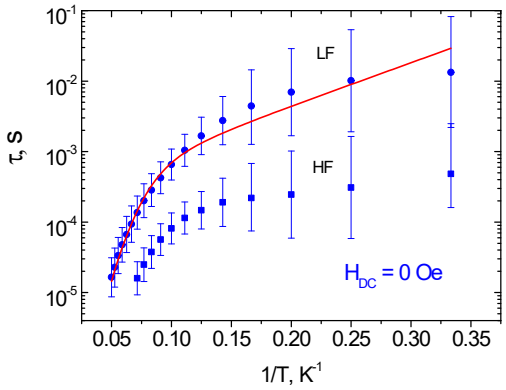
<td> 
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<td>Orbach+Raman
$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$

C_{Raman} = $5.27\cdot10^{-5} \pm 3.35\cdot10^{-6}$ $s^{-1}K^{-nRaman}$
n_{Raman} = 7 (fixed)
$\tau_0 = 2.65\cdot10^{-4} \pm 6.08\cdot10^{-5}$ s
Δ_{eff}/k_B = 14.1 ± 1.7 K
R^2 = 0.9667</td>
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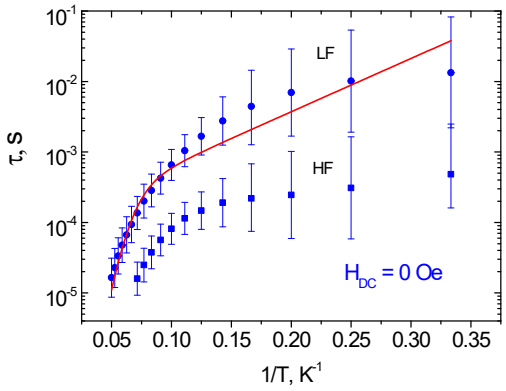
<td> 
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<td>Orbach+Raman
$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$

C_{Raman} = $1.71\cdot10^{-5} \pm 2.4\cdot10^{-6}$ $s^{-1}K^{-nRaman}$
n_{Raman} = 9 (fixed)
$\tau_0 = 1.15\cdot10^{-4} \pm 3.2\cdot10^{-5}$ s
Δ_{eff}/k_B = 17.4 ± 2.2 K
R^2 = 0.89321</td>
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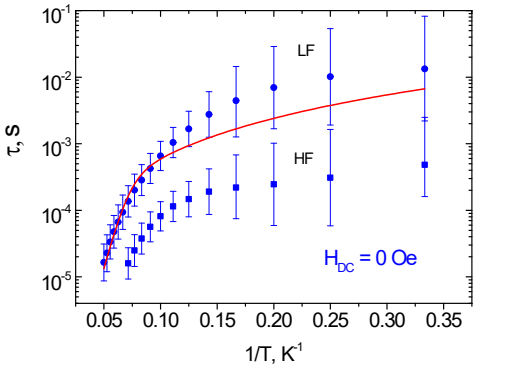
<td> 
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<td>Orbach+Rman
$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$

C_{Raman} = 16.6 ± 1.6 $s^{-1}K^{-nRaman}$
n_{Raman} = 2 (fixed)
$\tau_0 = 8.5\cdot10^{-9} \pm 1.14\cdot10^{-9}$ s
Δ_{eff}/k_B = 149 K(fixed)
R^2 = 0.90778</td>
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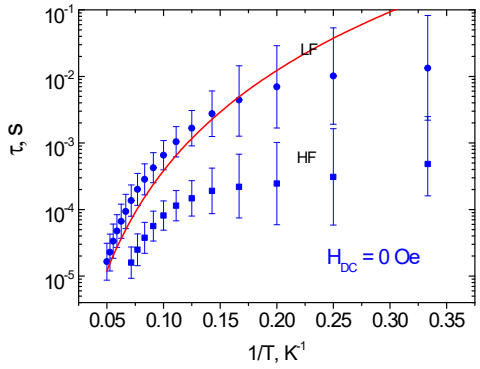
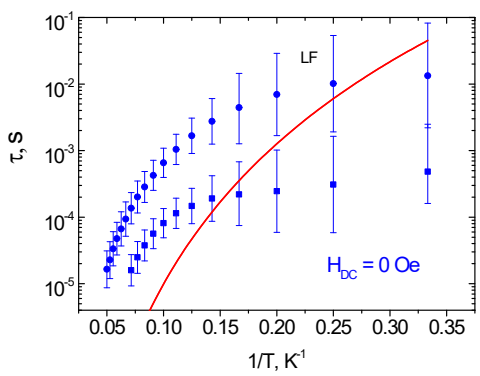
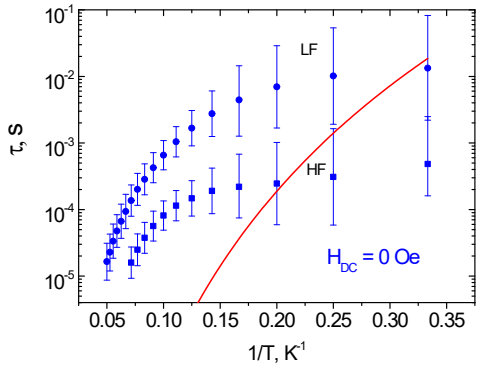
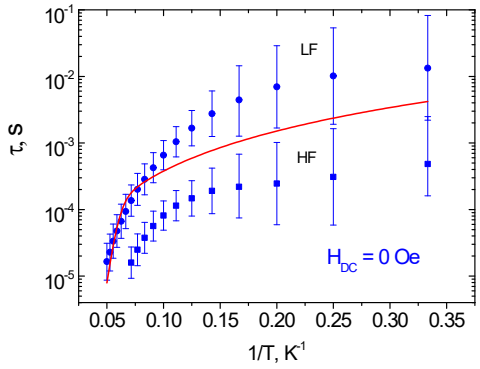
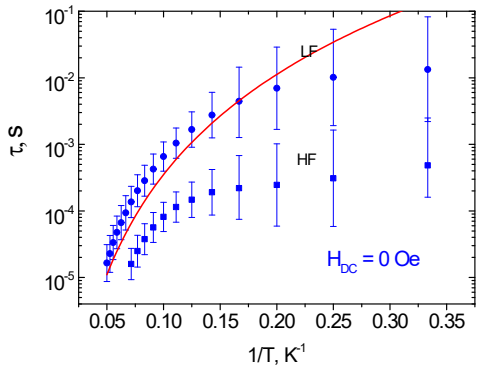
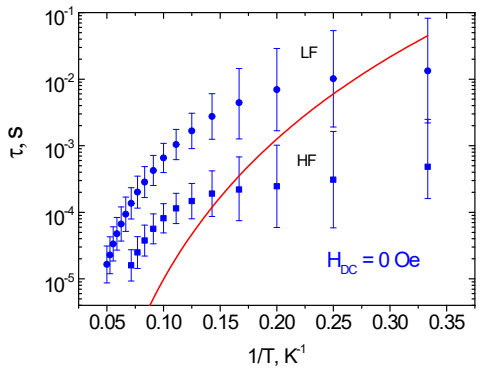
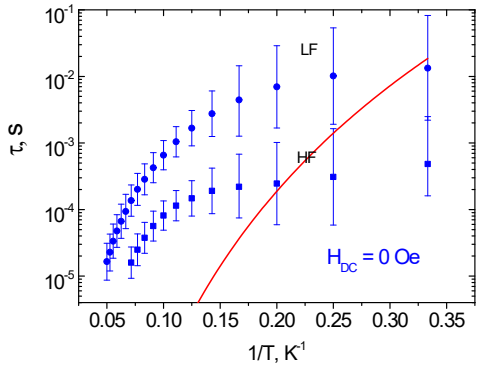
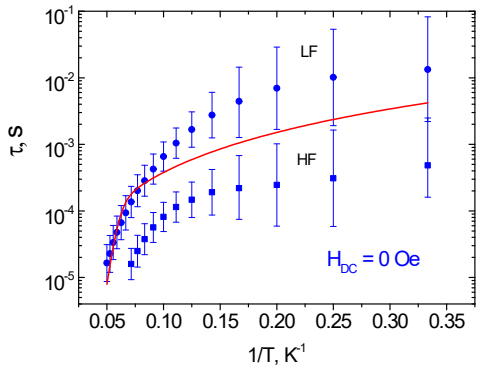
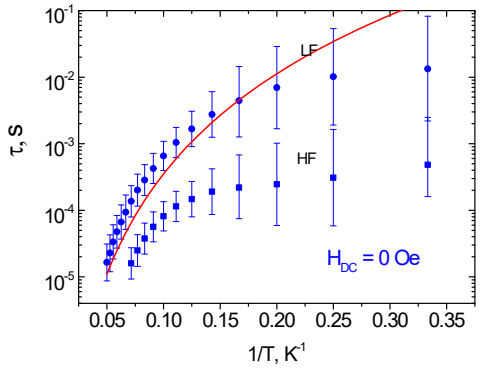
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ C_{Raman} = *0.026 ± 0.005* s^{-1}K$^{-nRaman}$ n_{Raman} = 5 (fixed) τ_0 = 8.74 s Δ_{eff}/k_B = 149 K(fixed) R^2 = 0.36919
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ C_{Raman} = *0.01013 ± 0.01812* s^{-1}K$^{-nRaman}$ n_{Raman} = 7 (fixed) τ_0 = 4.09·10^9 s Δ_{eff}/k_B = 149 K(fixed) R^2 = -0.65839
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ C_{Raman} = *0.00273 ± 0.0122* s^{-1}K$^{-nRaman}$ n_{Raman} = 9 (fixed) τ_0 = 1 ±·4.9·10^{15} s Δ_{eff}/k_B = 149 K(fixed) R^2 = -0.68581

	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = *26.5 ± 3.9* $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 1.5 \cdot 10^{-11} \pm 5.0 \cdot 10^{-12}$ s Δ_{eff}/k_B = 265 K(fixed) R^2 = 0.73765
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = *0.0285 ± 0.0065* $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) τ_0 = 0.43831 $\pm 8.7 \cdot 10^{9}$ s Δ_{eff}/k_B = 265 K(fixed) R^2 = 0.36187
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = *0.01018 ± 0.01829* $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) $\tau_0 = 1 \pm 9.6 \cdot 10^{14}$ s Δ_{eff}/k_B = 265 K(fixed) R^2 = -0.6584

	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = *0.00273* ± *0.0122* $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) τ_0 = 1 s Δ_{eff}/k_B = 265 K(fixed) R^2 = -0.68581
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = *26.5 ± 3.9* $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 1.5 \cdot 10^{-11} \pm 5.0 \cdot 10^{-12}$ s Δ_{eff}/k_B = 265 K(fixed) R^2 = 0.73765
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ C_{Raman} = *0.0285 ± 0.0065* $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) τ_0 = 0.43831 $\pm 8.7 \cdot 10^{9}$ s Δ_{eff}/k_B = 265 K(fixed) R^2 = 0.36187

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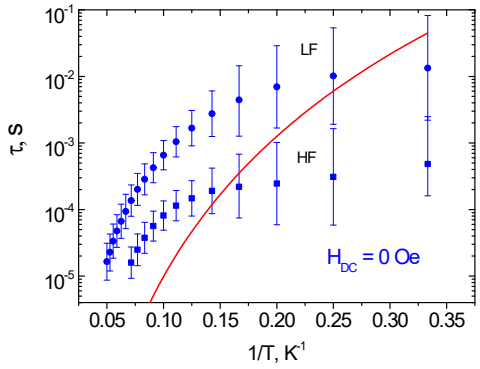
<td> 
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<td>Orbach+Raman
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$
C_{Raman} = 0.01018 ± 0.01829 $s^{-1}K^{-nRaman}$
n_{Raman} = 7 (fixed)
τ_0 = 1 ±·9.6·10¹⁴ s
Δ_{eff}/k_B = 265 K(fixed)
R^2 = -0.6584</td>
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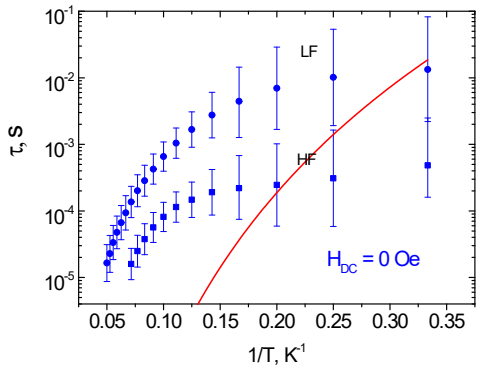
<td> 
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$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$
C_{Raman} = 0.00273 ± 0.0122 $s^{-1}K^{-nRaman}$
n_{Raman} = 9 (fixed)
τ_0 = 1 s
Δ_{eff}/k_B = 265 K (fixed)
R^2 = -0.68581</td>
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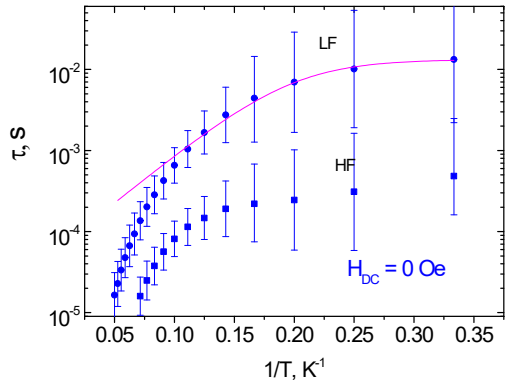
<td> 
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<td>QTM+Orbach
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + B$
B = 75.4 ± 2.04 s^{-1}
τ_0 = 5.8·10⁻⁵ ± 1.5·10⁻⁵ s
Δ_{eff}/k_B = 27.5 ± 1.6 K
R^2 = 0.99362</td>
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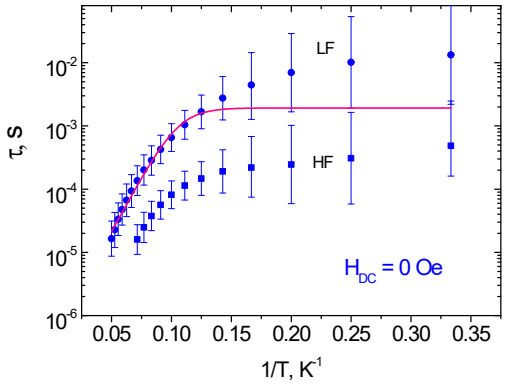
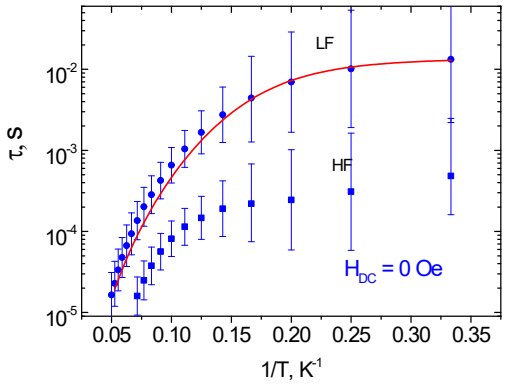
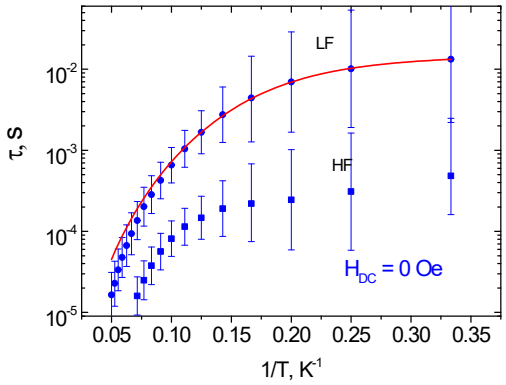
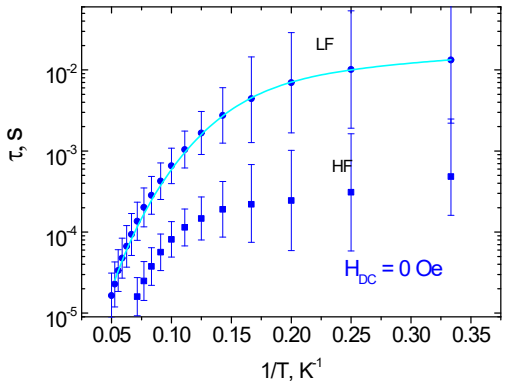
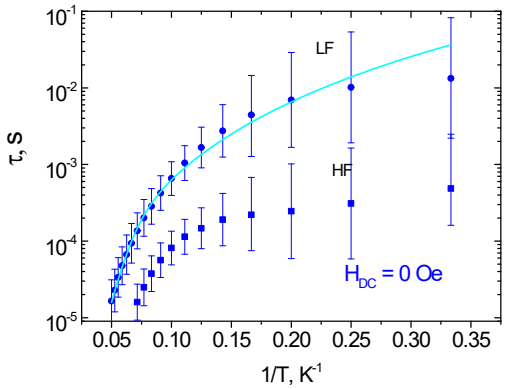
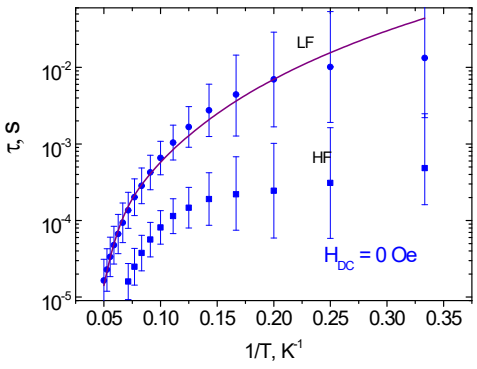
	**QTM+Orbach** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + B$ B = 520.4 ± 94.2 s^{-1} $\tau_0 = 3.79 \cdot 10^{-7} \pm 1.04 \cdot 10^{-7}$ s Δ_{eff}/k_B = 80 ± 4 K R^2 = 0.95044
	**QTM+Raman** $\tau^{-1} = C_{Raman} T^{nRaman} + B$ LF *B = 72.6 ± 1.5 s^{-1}* *C_{Raman} = 0.02 ± 0.001* $s^{-1}K^{-nRaman}$ *n_{Raman} = 5 (fixed)* *R^2 = 0.99587* We believe that this fit is not most plausible one because high-temperature ranges of zero-field τ *vs.* 1/T dependencesfor **1** (where the contribution of Orbach mechanism is reliably demonstrated) and **1_Y$_{0.5}$** are almost identical (Fig. 3e). Thus, it is plausible to involve Orbach mechanism to fit LF τ *vs.* 1/T dependency **1_Y$_{0.5}$**
	**QTM+Raman** $\tau^{-1} = C_{Raman} T^{nRaman} + B$ *B = 65.7 ± 0.8 s^{-1}* *C_{Raman} = 0.11 ± 0.014* $s^{-1}K^{-nRaman}$ *n_{Raman} = 4.06 ± 0.07* *R^2 = 0.99972* We believe that this fit is not most plausible one because high-temperature ranges of zero-field τ *vs.* 1/T dependencesfor **1** (where the contribution of Orbach mechanism is reliably demonstrated) and **1_Y$_{0.5}$** are almost

	identical (Fig. 3e). Thus, it is plausible to involve Orbach mechanism to fit LF τ *vs.* 1/T dependency **1_Y$_{0.5}$**
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = 0.016 ± 3.7·10^{-4} s^{-1}K$^{-nRaman}$ n_{Raman} = 5 (fixed) τ_0 = 7.34·10^{-3} ± 2.7·10^{-4} s Δ_{eff}/k_B = 2 ± 0.11 K *R^2 = 0.99976*
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = 0.68 ± 0.25 s^{-1}K$^{-nRaman}$ n_{Raman} = 3.36 ±0.16 τ_0 = 1.24·10^{-8} ± 1.47·10^{-9} s Δ_{eff}/k_B = 149 K (fixed) *R^2 = 0.97563*
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ C_{Raman} = 0.44 ± 0.176 s^{-1}K$^{-nRaman}$ n_{Raman} = 3.6 ± 0.17 τ_0 = 9.6·10^{-10} ± 1.8·10^{-10} s Δ_{eff}/k_B = 199 K (fixed) *R^2 = 0.95595*

<table>
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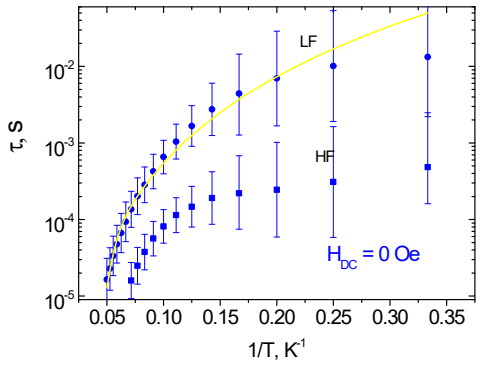
<td> 
</td>
<td>Raman+Orbach
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman}$

C_{Raman} = 0.33 ± 0.14 $s^{-1}K^{-nRaman}$
N_{Raman} = 3.75 ± 0.18
$\tau_0 = 3.4 \cdot 10^{-11} \pm 8.9 \cdot 10^{-12}$ s
Δ_{eff}/k_B = 265 K (fixed)
$R^2 = 0.93459$</td>
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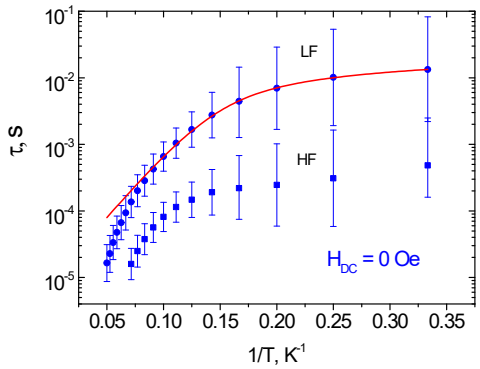
<td> 
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<td>Orbach+Raman+QTM
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$

B = 44.3 ± 1.5 s^{-1}
C_{Raman} = 3.4 ± 0.14 $s^{-1}K^{-nRaman}$
n_{Raman} = 2 (fixed)
$\tau_0 = 9.2 \cdot 10^{-6} \pm 2.1 \cdot 10^{-6}$ s
Δ_{eff}/k_B = 45.4 ± 1.8 K
R^2 = 0.99959</td>
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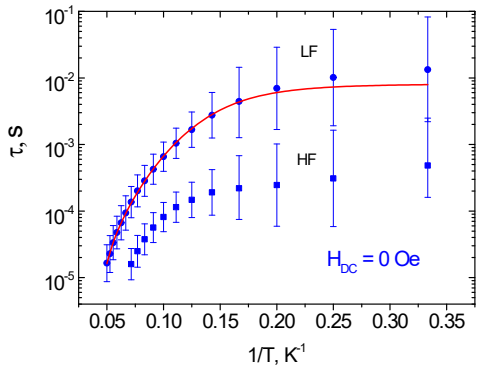
<td> 
</td>
<td>Orbach+Raman+QTM
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman} T^{nRaman} + B$

B = 123 ± 11 s^{-1}
C_{Raman} = 0.0138 ± $2.06 \cdot 10^{-4}$ $s^{-1}K^{-nRaman}$
n_{Raman} = 5 (fixed)
$\tau_0 = 4.5 \cdot 10^{-11} \pm 1.1 \cdot 10^{-10}$ s
Δ_{eff}/k_B = 281 ± 49.3 K
R^2 = 0.99789</td>
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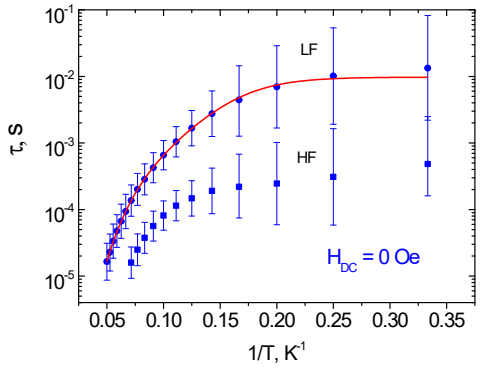
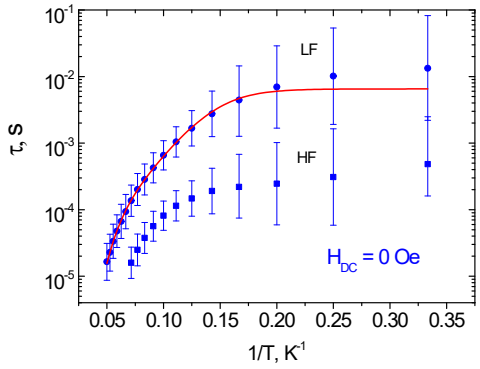
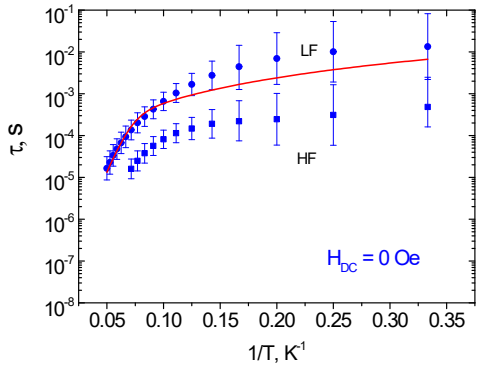
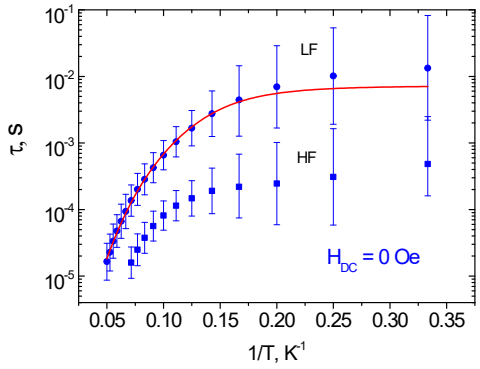
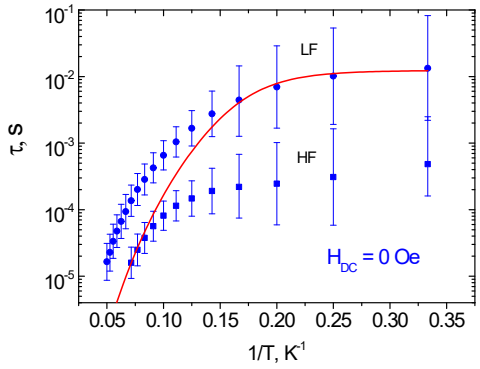
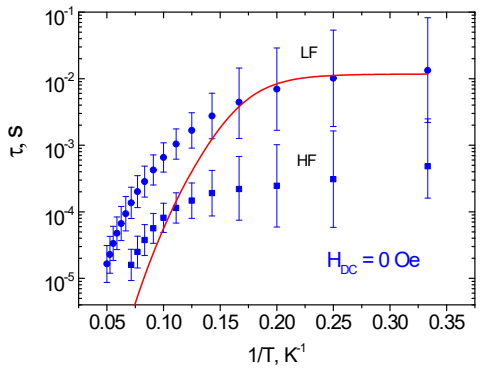
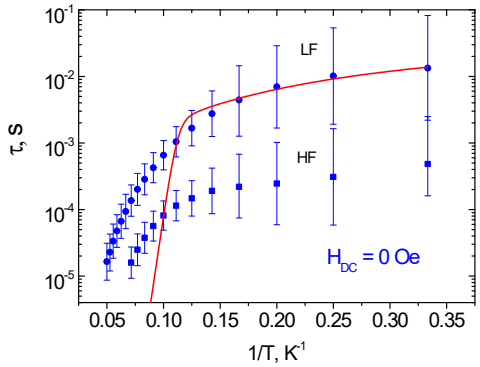
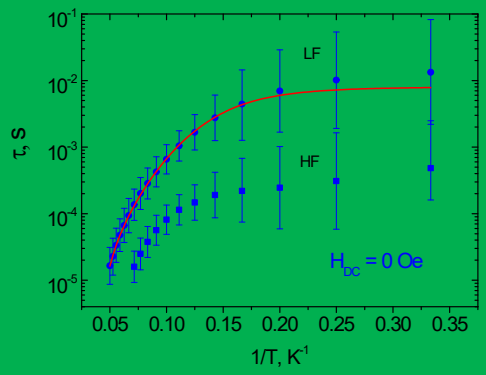
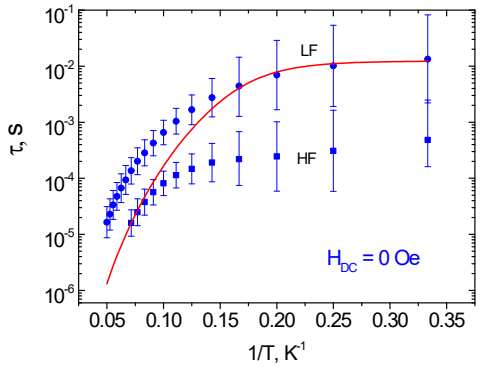
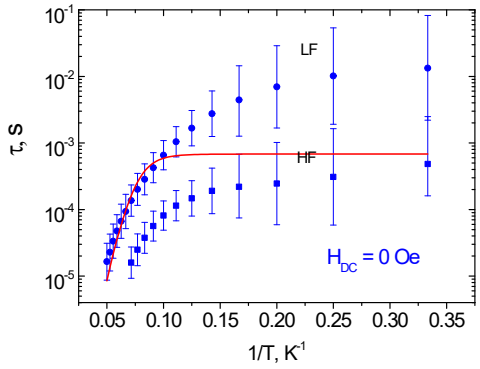
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}+B$ B = 102.6 ± 9.2 s^{-1} C_{Raman} = $4.18\cdot10^{-5}$± $8.4\cdot10^{-7}$ $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) $\tau_0 = 2.8\cdot10^{-5} \pm 3.2\cdot10^{-6}$ s Δ_{eff}/k_B = 35.6 ± 1.1 K R^2 = 0.99897
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}+B$ B = 135.5 ± 18.95 s^{-1} C_{Raman} = $9.85\cdot10^{-8}$± $4.94\cdot10^{-9}$ $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) $\tau_0 = 7.2\cdot10^{-6} \pm 1.01\cdot10^{-6}$ s Δ_{eff}/k_B = 46.5 ± 1.5 K R^2 = 0.99667
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + +C_{Raman}T^{nRaman}+B$ B = 0 ± 336 s^{-1} C_{Raman} = 16.7 ± 3.6 $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 8.5\cdot10^{-9} \pm 1.3\cdot10^{-9}$ s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.90163

	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + + C_{Raman} T^{nRaman} + B$ B = 138.7 ± 19.9 s^{-1} C_{Raman} = 0.01322 ± 3.87·10^{-4} $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) τ_0 = 4.6·10^{-8} ± 9.3·10^{-9} s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.99417
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + + C_{Raman} T^{nRaman} + B$ B = 80.55 ± 3.45 s^{-1} C_{Raman} = 5.96·10^{-4} ± 9.38·10^{-5} $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) τ_0 = 1 ± 2.9·10^{9} s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.97454
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + + C_{Raman} T^{nRaman} + B$ B = 84.9 ± 4.8 s^{-1} C_{Raman} = 1.8·10^{-5} ± 4.6·10^{-6} $s^{-1}K^{-nRaman}$ n_{Raman} = 9 (fixed) τ_0 = 1 ± 3.56·10^{11} s Δ_{eff}/k_B = 149 K (fixed) R^2 = 0.95101

	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + + C_{Raman} T^{nRaman} + B$ B = 25.8 ± 5.5 s^{-1} C_{Raman} = 5.22 ± 0.45 $s^{-1}K^{-nRaman}$ n_{Raman} = 2 (fixed) $\tau_0 = 2.54 \cdot 10^{-16} \pm 2.32 \cdot 10^{-16}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.98335
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + + C_{Raman} T^{nRaman} + B$ B = 124.3 ± 10.2 s^{-1} C_{Raman} = 0.0137 ± 1.74·10^{-4} $s^{-1}K^{-nRaman}$ n_{Raman} = 5 (fixed) $\tau_0 = 1.02 \cdot 10^{-10} \pm 1.13 \cdot 10^{-11}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.99800
	**Orbach+Raman+QTM** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + + C_{Raman} T^{nRaman} + B$ B = 80.6 ± 3.4 s^{-1} C_{Raman} = 5.96·10^{-4} ± 9.29·10^{-5} $s^{-1}K^{-nRaman}$ n_{Raman} = 7 (fixed) $\tau_0 = 1 \pm 8.2 \cdot 10^{13}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.97454



Orbach+Raman+QTM

$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + +C_{Raman}T^{nRaman}+B$

B = 1467 ± 3747 s^{-1}

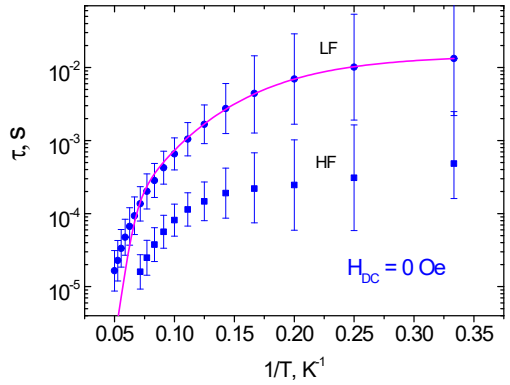
C_{Raman} = $2.27\cdot10^{-7}$ ± $3.95\cdot10^{-6}$ $s^{-1}K^{-nRaman}$

n_{Raman} = 9 (fixed)

τ_0 = 0.52 ± $3\cdot10^{12}$ s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = -0.33725



QTM+Raman+Orbach

$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B+C_{Raman}T^{nRaman}$

B = 65 ± 0.7 s^{-1}

C_{Raman} = *0.12 ± 0.014* $s^{-1}K^{-nRaman}$

N_{Raman} = *4.03 ± 0.06*

τ_0 = $3.3\cdot10^{-12}$ ± $3.4\cdot10^{-12}$ s

Δ_{eff}/k_B = 265 K (fixed)

R^2 = 0.99972

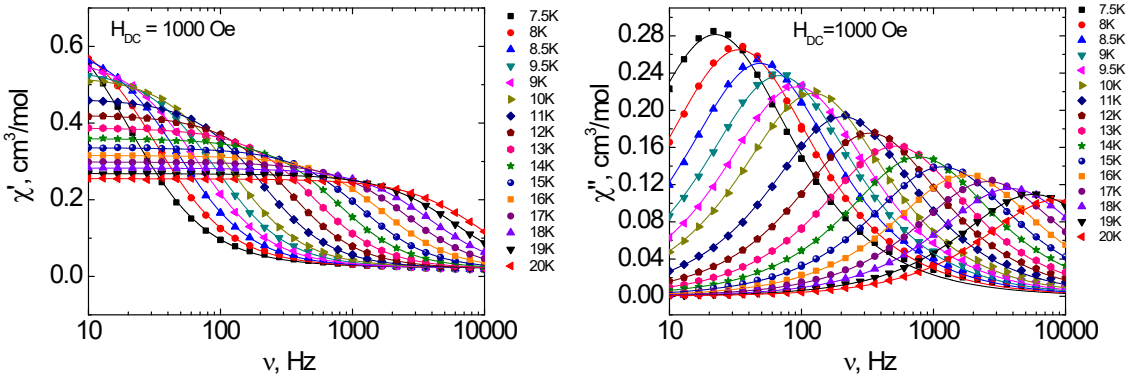


Fig. S24. Frequency dependencies of real (χ', left) and imaginary (χ", right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.5}$** in 1000 Oe magnetic field. Solid lines shows approximations of experimental data by the generalized Debye model.

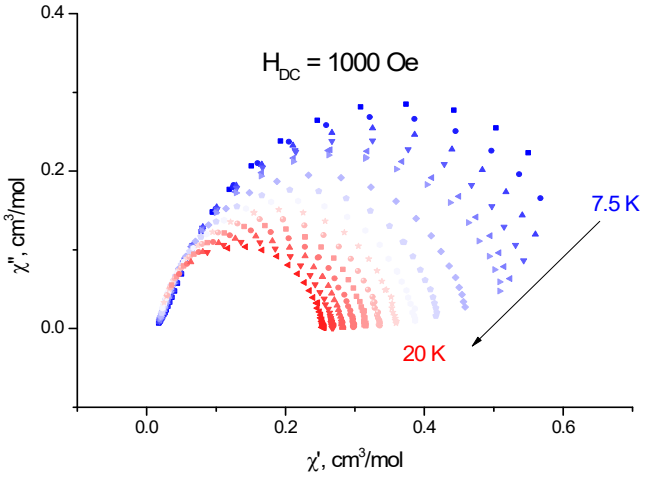
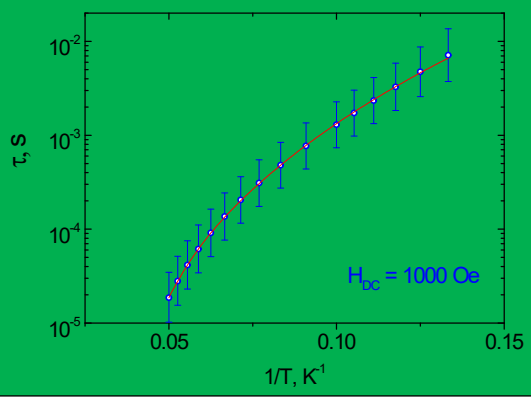
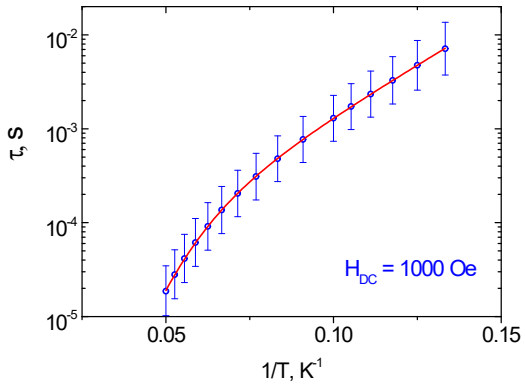
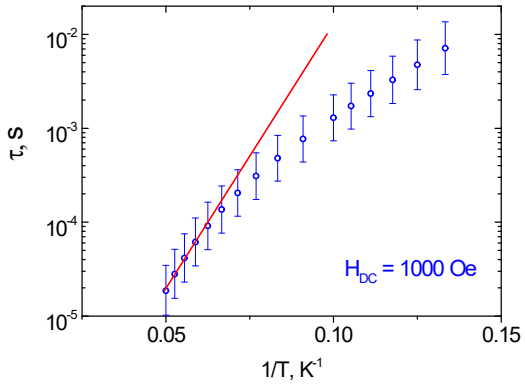


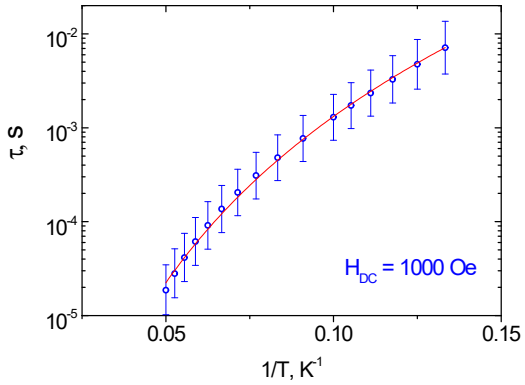
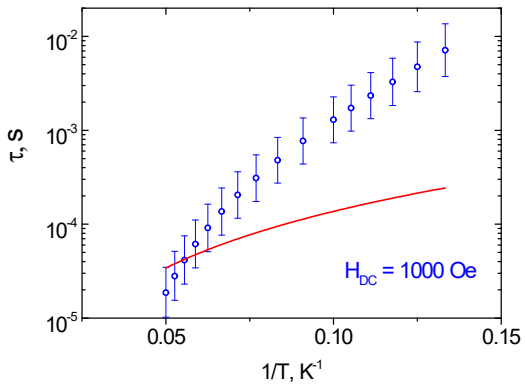
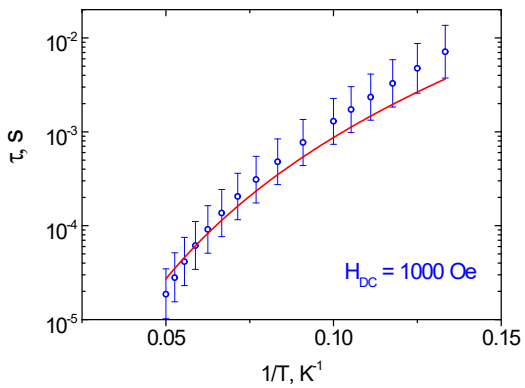
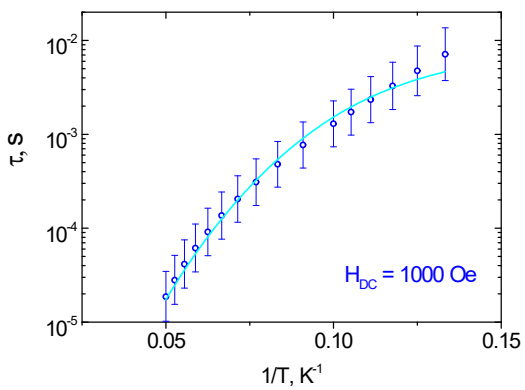
Fig. S25. Cole-Cole plots for **1_Y$_{0.5}$** in optimal DC field of 1000 Oe.

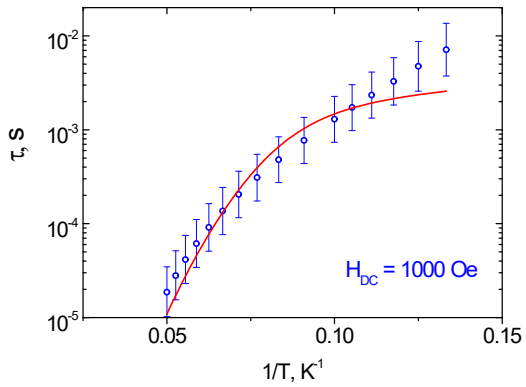
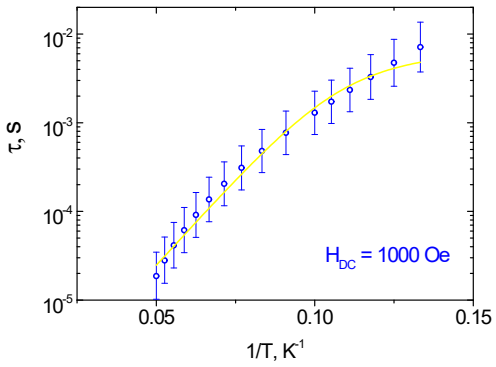
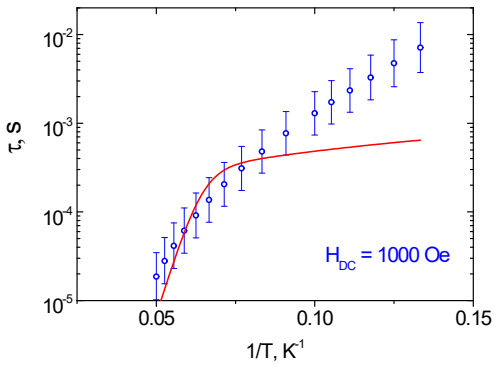
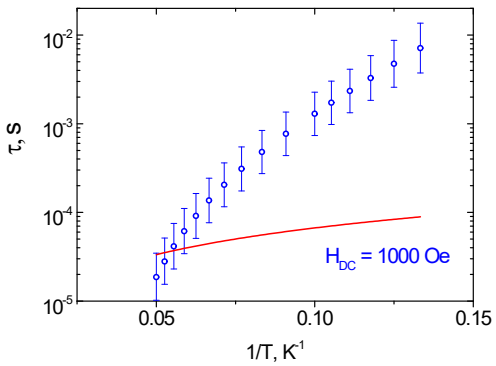
Table. S12. Distribution of parameter α at H_{DC} = 1000 Oe for **1_Y$_{0.5}$**.

T, K	α
7.5	0.11751
8	0.10652
8.5	0.09897
9	0.09386
9.5	0.08947
10	0.08889
11	0.0864
12	0.08502
13	0.08474
14	0.08505
15	0.08493
16	0.08728
17	0.08326
18	0.08698
19	0.09009
20	0.09726

Table S13. Fitting of the τ *vs. 1/T* dependence for **1_Y$_{0.5}$** (H_{DC} = 1000 Oe, T = 7.5-20 K). Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color.

Fitting line	**Mechanism(s)** Parameters
	**Raman+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+C_{Raman}T^{nRaman}$ $C_{Raman} = 0.0020\pm1\cdot10^{-4}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 5.59\pm0.03$ $\tau_0 = 1.00\cdot10^{-10}\pm9\cdot10^{-12}$ s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.99923
	**Raman+Orbach** $C_{Raman} = 5.72\cdot10^{-7}\pm2.75\cdot10^{-7}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 8.4\pm0.2$ $\tau_0 = 1.2\cdot10^{-5}\pm7.04\cdot10^{-7}$ s $\Delta_{eff}/k_B = 48.7\pm0.4$ K R^2 = 0.99989
	**Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)$ $\tau_0 = 3.0\cdot10^{-8} \pm 8\cdot10^{-9}$ s $\Delta_{eff}/k_B = 130 \pm 5$ K R^2 = 0.99651

	**Direct+Raman** $\tau^{-1} = C_{Raman}T^{nRaman}+A_{direct}TH^4$ C_{Raman}= $1\cdot10^{-3}\pm 1.49\cdot10^{-3}$ $s^{-1}K^{-nRaman}$ n_{Raman}= 5.88 ± 0.16 $A_{direct} = 0\pm 1.7\cdot10^{-8}$ $K^{-1}Oe^{-4}s^{-1}$ R^2 = 0.99003
	**Direct+Raman** $\tau^{-1} = C_{Raman}T^{nRaman}+A_{direct}TH^4$ C_{Raman}= 73± 2643 $s^{-1}K^{-nRaman}$ n_{Raman}=2(fixed) $A_{direct} = 0\pm 6\cdot10^{-7}$ $K^{-1}Oe^{-4}s^{-1}$ R^2 = 0.13834
	**Direct+Raman** $\tau^{-1} = C_{Raman}T^{nRaman}+A_{direct}TH^4$ C_{Raman}= 0.01 ± 0.03 $s^{-1}K^{-nRaman}$ n_{Raman}=5(fixed) $A_{direct} = 0\pm 3.8\cdot10^{-8}$ $K^{-1}Oe^{-4}s^{-1}$ R^2 = 0.85633
	**Direct+Raman** $\tau^{-1} = C_{Raman}T^{nRaman}+A_{direct}TH^4$ C_{Raman}= $4.48\cdot10^{-5}\pm 2.5\cdot10^{-6}$ $s^{-1}K^{-nRaman}$ n_{Raman}=7(fixed) $A_{direct} = 2.08\cdot10^{-11}\pm 3\cdot10^{-12}$ $K^{-1}Oe^{-4}s^{-1}$ R^2 = 0.97015

	**Direct+Raman** $\tau^{-1} = C_{\text{Raman}}T^{\text{nRaman}}+A_{direct}TH^4$ C_{Raman}= 1.77·10-7± 2.7·10-8 s-1K-nRaman n_{Raman}=9(fixed) A_{direct} = ·5·10-11± 1·10-11 K-1Oe-4s-1 R^2 = 0.84961
	**Direct+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+A_{direct}TH^4$ A_{direct} = 2.49·10-11 ± 4.6·10-12 K-1Oe-4s-1 τ_0 = 2.7·10-7±7.8·10-8 s Δ_{eff}/k_B =90.7 ± 4.4 K R^2 = 0.9565
	**Direct+Orbach** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{\text{eff}}/k_BT)+A_{\text{direct}}TH^4$ A_{direct} = 2.0·10-10 ± 6·10-11 K-1Oe-4s-1 τ_0 = 1.2·10-11±6·10-12 s Δ_{eff}/k_B = 265 K (fixed) R^2 = 0.50456
	**Direct** $\tau^{-1} = A_{direct}TH^4$ A_{direct} = 1495.8 ± 517.9 K-1Oe-4s-1 R^2 = 0.07981

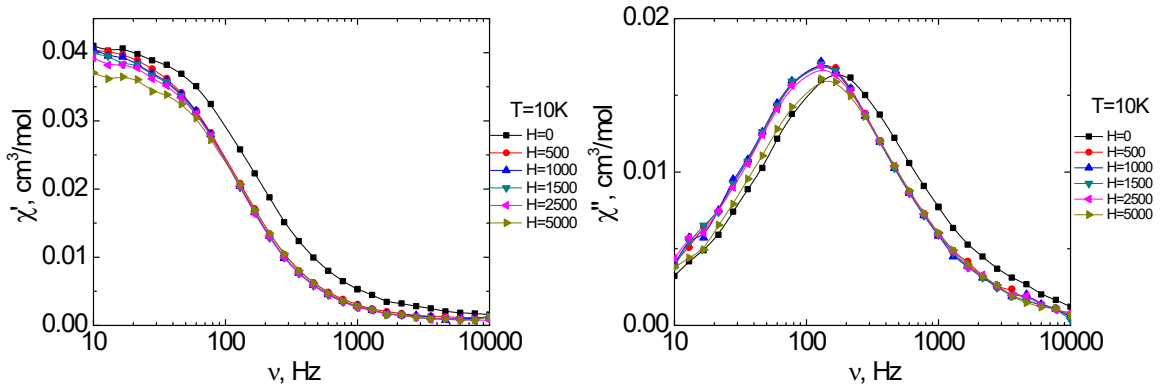


Fig. S26. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ'', right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.9}$** in various DC magnetic fields at 10 K. Solid lines are visual guides.

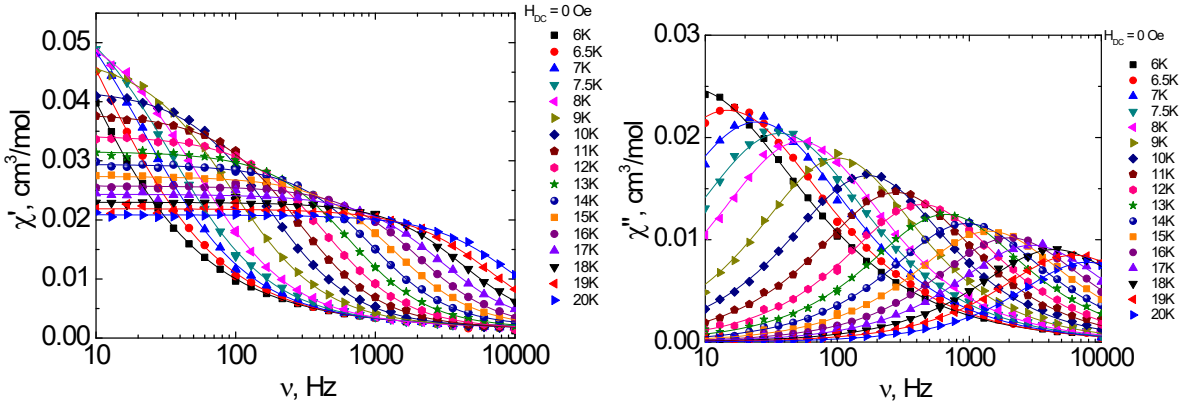


Fig. S27. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ'', right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.9}$** in zero DC magnetic field. Solid lines shows approximations of experimental data by the generalized Debye model.

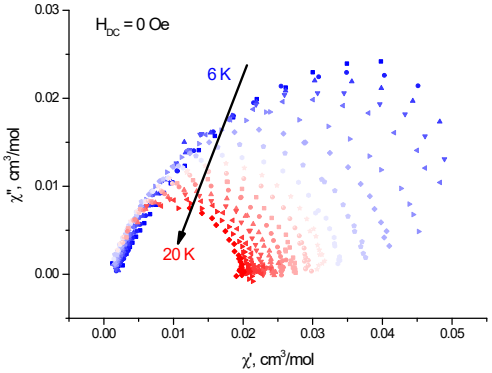
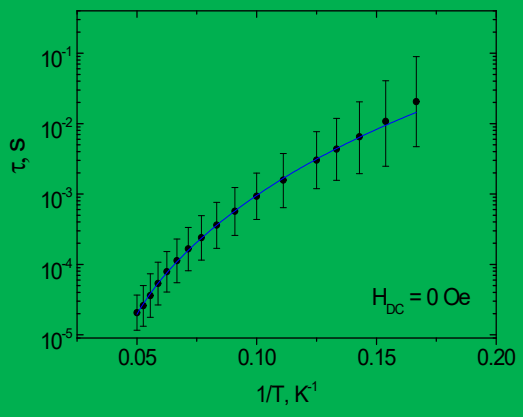
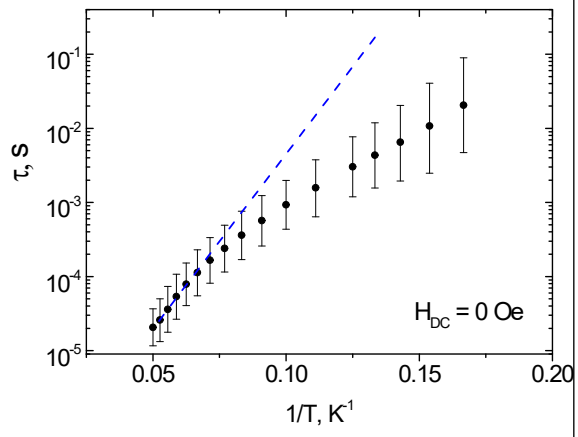
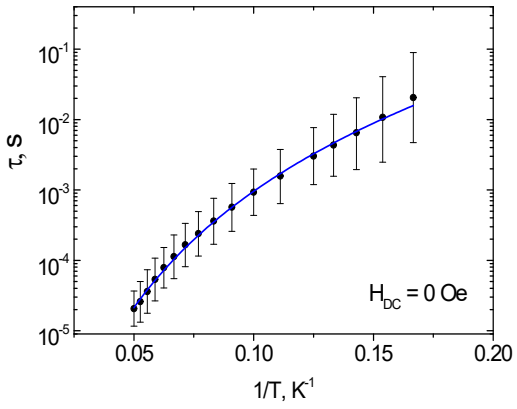


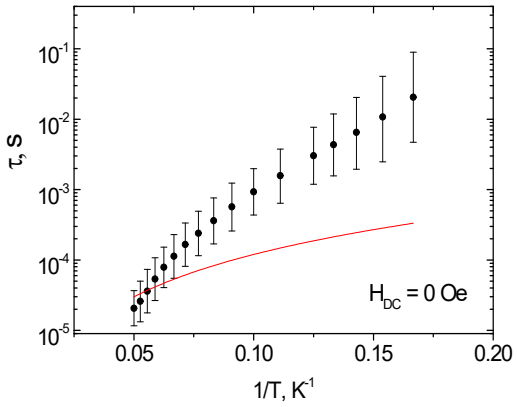
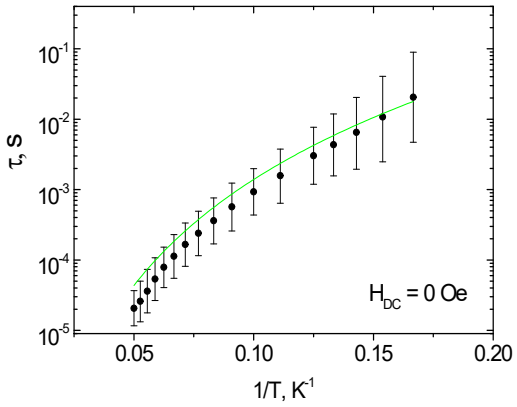
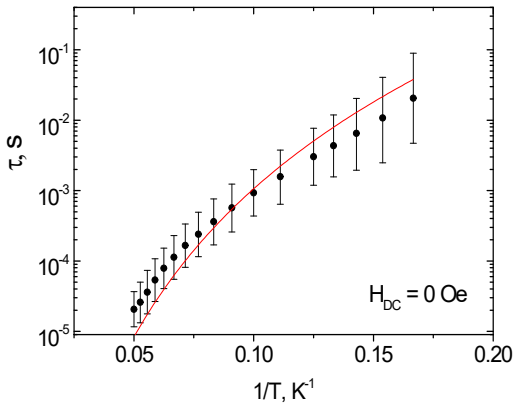
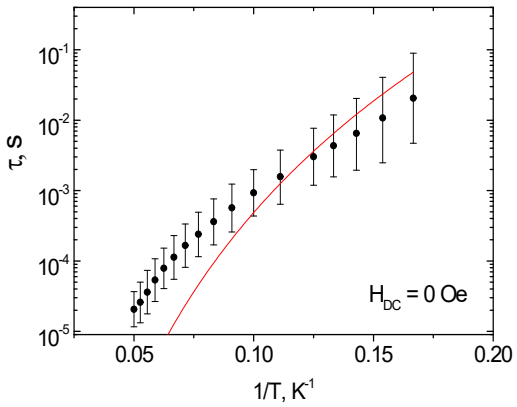
Fig. S28. Cole-Cole plots for **1_Y$_{0.9}$** in zero DC field.

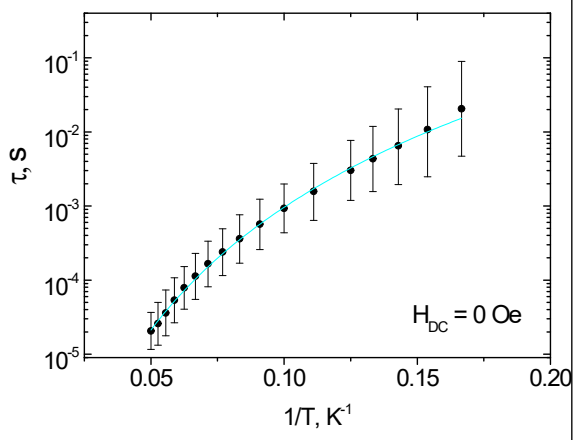
Table. S14. Distribution of parameter α at H_{DC} = 0 Oe for **1_Y$_{0.9}$**.

T, K	α
6	0.31912
6.5	0.26603
7	0.22886
7.5	0.20502
8	0.18463
9	0.14642
10	0.14593
11	0.14116
12	0.13106
13	0.13104
14	0.12882
15	0.12597
16	0.11336
17	0.11817
18	0.1237
19	0.11247
20	0.07005

Table S15. Fitting of the τ *vs. 1/T* dependence for **1_Y$_{0.9}$**(H_{DC} = 0 Oe, T = 6-20 K). Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color.

Fitting line	**Mechanism(s)** Parameters
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 265 K (fixed) $\tau_0 = 1.9\cdot10^{-10} \pm 5.7\cdot10^{-11}$ s $C_{Raman} = 0.0049 \pm 8.04\cdot10^{-4}$ s^{-1}K$^{-nRaman}$ $n_{Raman} = 5.3 \pm 0.1$ **R^2 = 0.99493**
	**Orbach** $\tau = \tau_0\cdot\exp(\Delta_{eff}/k_BT)$ T = 6-7 K Δ_{eff}/k_B = 103.8 ± 7.5 K $\tau_0 = 1.1\cdot10^{-7} \pm 4.5\cdot10^{-8}$s R^2 = 0.99031
	**Raman** $\tau^{-1} = C_{Raman}T^{n\_Raman}$ $C_{Raman} = 0.0034 \pm 4.9\cdot10^{-4}$s^{-1}K$^{-nRaman}$ $n_{Raman} = 5.49 \pm 0.06$ R^2 = 0.99133

	**Raman** $\tau^{-1} = C_{Raman} T^{n\_Raman}$ C_{Raman}=83.5± 16.5 $s^{-1}K^{-nRaman}$ n_{Raman}=2 (fixed) R^2 = 0.26684
	**Raman** $\tau^{-1} = C_{Raman} T^{n\_Raman}$ C_{Raman}=0.0072 ± $3 \cdot 10^{-4} s^{-1}K^{-nRaman}$ n_{Raman}=5 (fixed) R^2 = 0.96544
	**Raman** $\tau^{-1} = C_{Raman} T^{n\_Raman}$ C_{Raman}=$9.4 \cdot 10^{-5}$± $1.3 \cdot 10^{-5} s^{-1}K^{-nRaman}$ n_{Raman}=7 (fixed) R^2 = 0.53346
	**Raman** $\tau^{-1} = C_{Raman} T^{n\_Raman}$ C_{Raman}=$2.06 \cdot 10^{-6}$± $7.6 \cdot 10^{-7} s^{-1}K^{-nRaman}$ n_{Raman}=9 (fixed) R^2 = -0.30966



Raman+ QTM

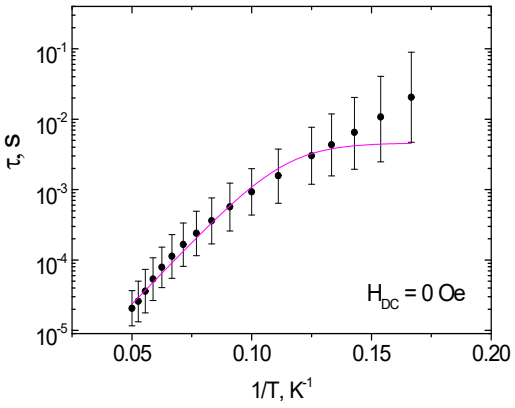
$\tau^{-1} = C_{Raman}T^{nRaman} + B$

C_{Raman}= 0.0033 ± 7.6·10^{-4} s^{-1}K$^{-nRaman}$

n_{Raman}= 5.5 ± 0.09

B = 2.9± 14.06 s^{-1}

R^2 = 0.99073



Orbach+QTM

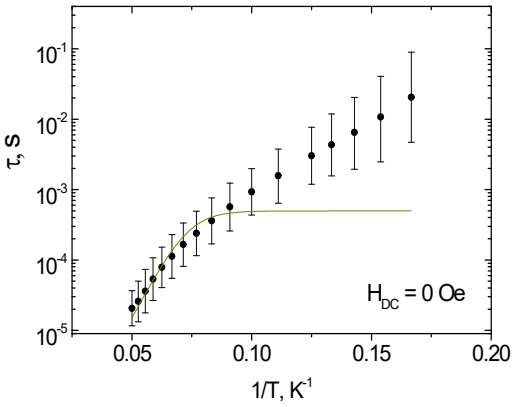
$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B$

Δ_{eff}/k_B = 82±3.7 K

τ_0=4·10^{-7}± 9.6·10^{-8} s

B= 218 ± 55 s^{-1}

R^2 = 0.93275



Orbach+QTM

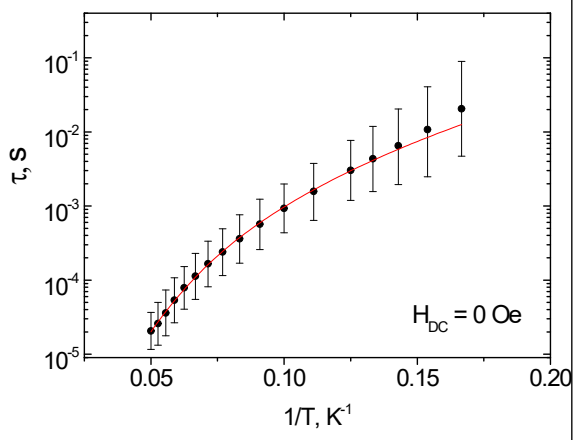
$\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)+B$

Δ_{eff}/k_B = 149 K (fixed)

τ_0=9·10^{-9}± 1.5·10^{-9} s

B= 2011 ± 492 s^{-1}

R^2 = 0.71506



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$

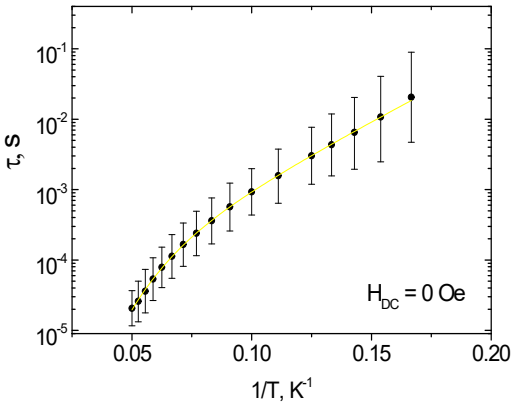
Δ_{eff}/k_B = 149 K (fixed)

$\tau_0 = 3.4 \cdot 10^{-8} \pm 3.3 \cdot 10^{-9}$ s

$C_{Raman} = 0.01 \pm 2.37 \cdot 10^{-4}$ $s^{-1}K^{-nRaman}$

$n_{Raman} = 5$ (fixed)

$R^2 = 0.99384$



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$

$\Delta_{eff}/k_B = 42 \pm 1.4$ K

$\tau_0 = 2.1 \cdot 10^{-5} \pm 5.2 \cdot 10^{-6}$ s

$C_{Raman} = 3.78 \cdot 10^{-5} \pm 4.15 \cdot 10^{-5}$ $s^{-1}K^{-nRaman}$

$n_{Raman} = 6.98 \pm 0.38$

$R^2 = 0.99782$

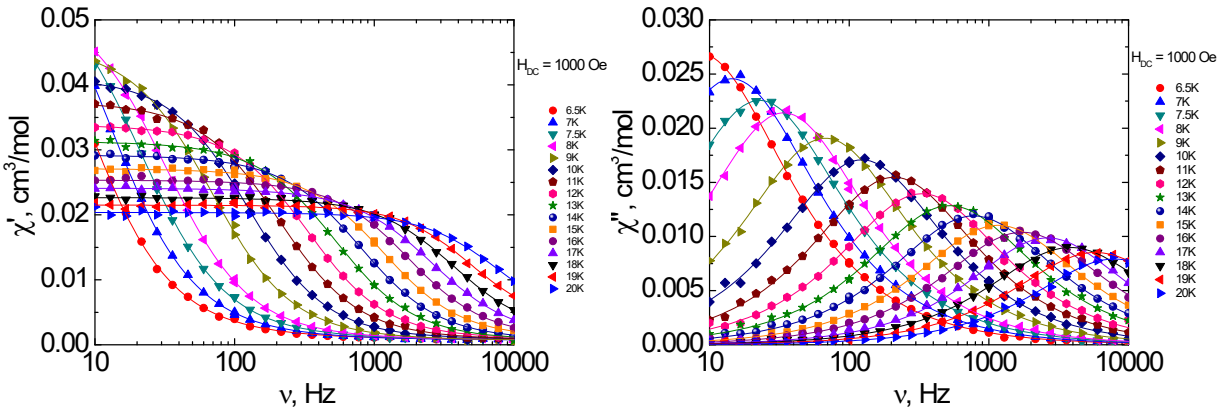


Fig. S29. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ'', right) components of dynamic magnetic susceptibility of complex **1_$Y_{0.9}$** in optimal DC magnetic field of 1000 Oe. Solid lines show approximations of experimental data by the generalized Debye model.

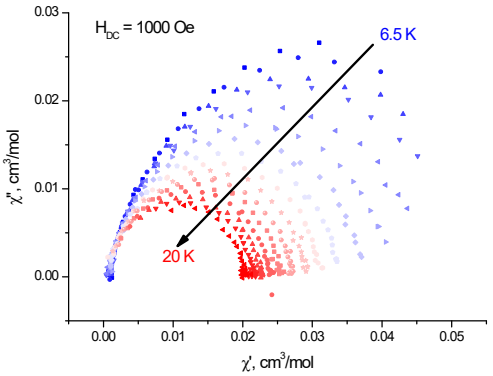
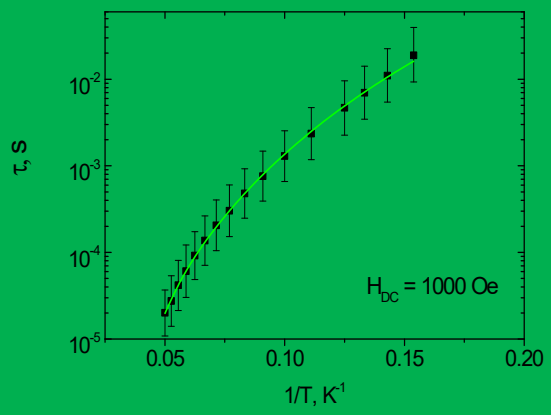
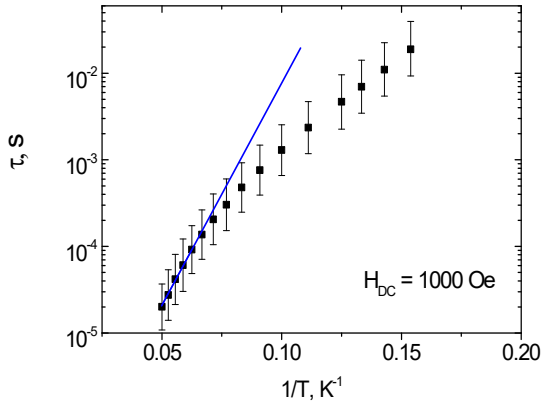
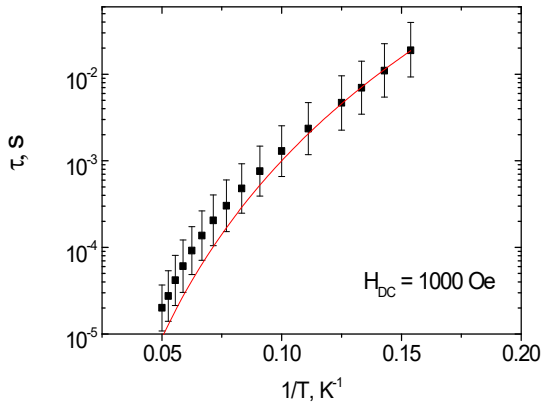


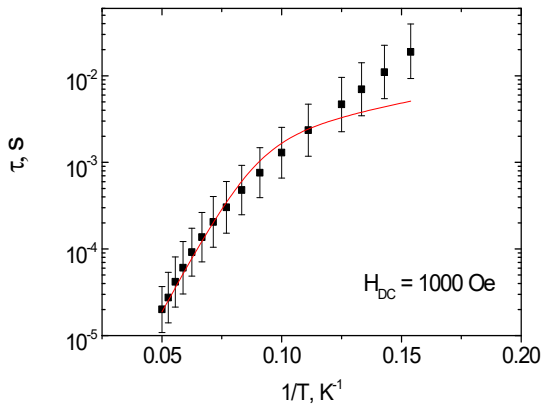
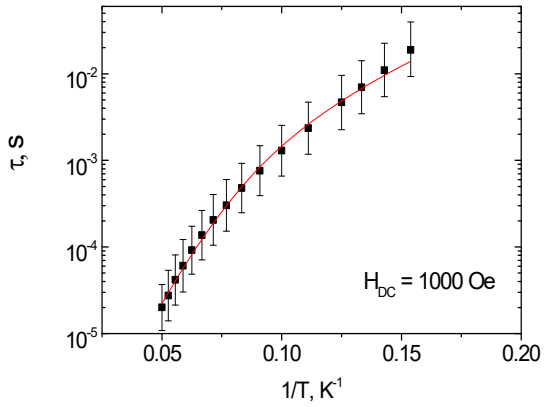
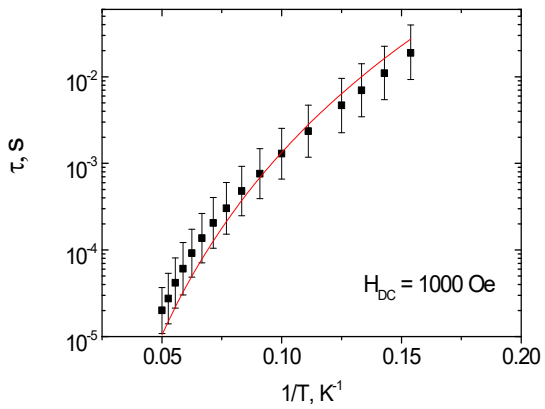
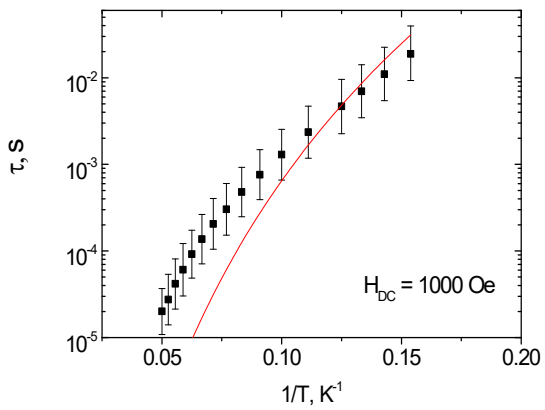
Fig. S30. Cole-Cole plots for **1_Y$_{0.9}$** in optimal DC field of 1000 Oe.

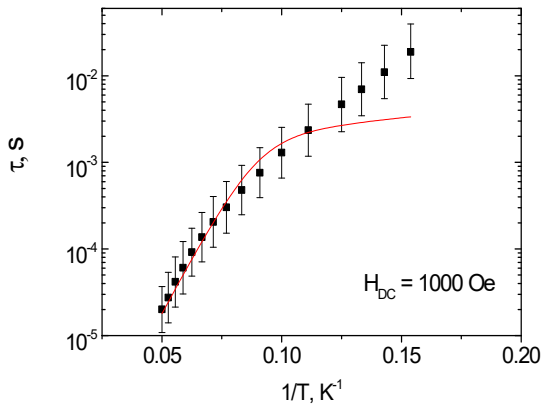
Table. S16. Distribution of parameter α at H_{DC} = 1000 Oe for **1_Y$_{0.9}$**.

T, K	α
6.5	0.13717
7	0.12143
7.5	0.11714
8	0.11773
9	0.11383
10	0.10964
11	0.10649
12	0.10455
13	0.11185
14	0.11294
15	0.10656
16	0.10092
17	0.11423
18	0.10416
19	0.10788
20	0.08895

Table S17. Fitting of the τ *vs. 1/T* dependence for **1_Y$_{0.9}$**(H_{DC} = 1000 Oe, T = 6.5-20 K). Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color.

Fitting line	**Mechanism(s)** Parameters
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 265 K (fixed) $\tau_0 = 1.5\cdot10^{-10} \pm 3.6\cdot10^{-11}$ s $C_{Raman} = 0.00134 \pm 1.8\cdot10^{-4}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 5.73 \pm 0.06$ $R^2 = 0.99622$
	***Orbach*** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT)$ $\Delta_{eff}/k_B = 118.6 \pm 3$ K $\tau_0 = 5.5\cdot10^{-8} \pm 1\cdot10^{-10}$ s $R^2 = 0.9981$
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1}\cdot\exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ $\Delta_{eff}/k_B = 118.6$ K *(fixed)* $\tau_0 = 1 \pm 2.33\cdot10^{7}$ s $C_{Raman} = 1.6\cdot10^{-4} \pm 7\cdot10^{-5}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 6.79 \pm 0.23$ $R^2 = 0.99879$

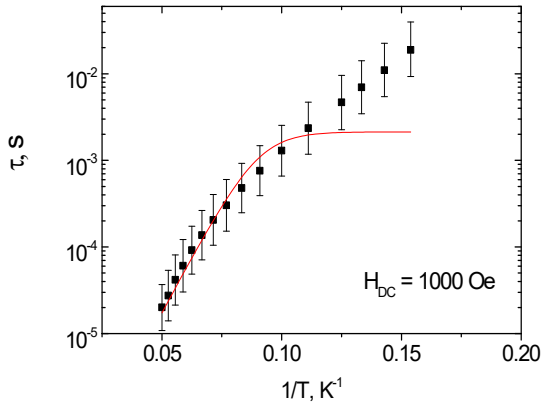
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118.6 K (fixed) $\tau_0 = 5.2 \cdot 10^{-8} \pm 7 \cdot 10^{-9}$ s C_{Raman}= 4.6± 0.8 $s^{-1}K^{-nRaman}$ n_{Raman}= 2 (fixed) R^2 = 0.85771
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118.6 K (fixed) $\tau_0 = 1 \cdot 10^{-7} \pm 1.0 \cdot 10^{-8}$ s C_{Raman}= $6.19 \cdot 10^{-3} \pm 2.9 \cdot 10^{-4}$ $s^{-1}K^{-nRaman}$ n_{Raman}= 5 (fixed) R^2 = 0.98295
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118.6 K (fixed) $\tau_0 = 1 \pm 1.05 \cdot 10^{7}$ s C_{Raman}= $7.5 \cdot 10^{-5} \pm 1.1 \cdot 10^{-5}$ $s^{-1}K^{-nRaman}$ n_{Raman}= 7 (fixed) R^2 = 0.75921
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118.6 K (fixed) $\tau_0 = 1 \pm 2.1 \cdot 10^{8}$ s C_{Raman}= $1.55 \cdot 10^{-6} \pm 6.3 \cdot 10^{-7}$ $s^{-1}K^{-nRaman}$ n_{Raman}= 9 (fixed) R^2 = 0.10822



Orbach+Direct

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + A_{direct} T H^4$

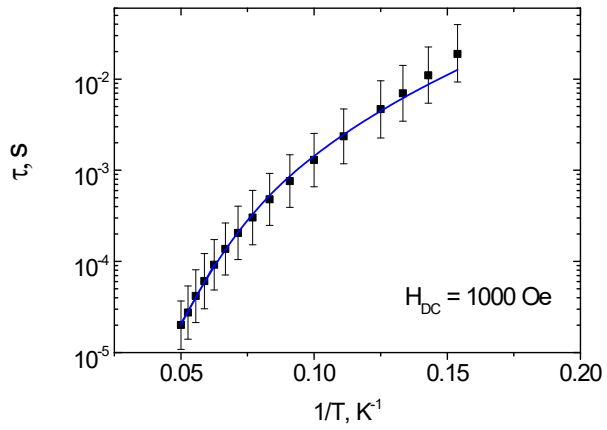
Δ_{eff}/k_B = *118.6 K (fixed)*
$\tau_0 = 4.8 \cdot 10^{-8} \pm 7.2 \cdot 10^{-9}$ *s*
$A_{direct} = 4.6 \cdot 10^{-11} \pm 1.1 \cdot 10^{-11}$ $K^{-1}Oe^{-4}s^{-1}$
$R^2 = 0.80075$



Orbach+QTM

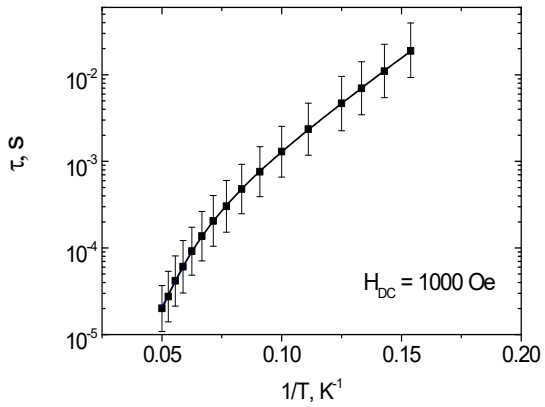
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + B$

Δ_{eff}/k_B = *118.6 K (fixed)*
$\tau_0 = 4.7 \cdot 10^{-8} \pm 7 \cdot 10^{-9}$ *s*
$B = 468.6 \pm 134.3$ s^{-1}
$R^2 = 0.74693$



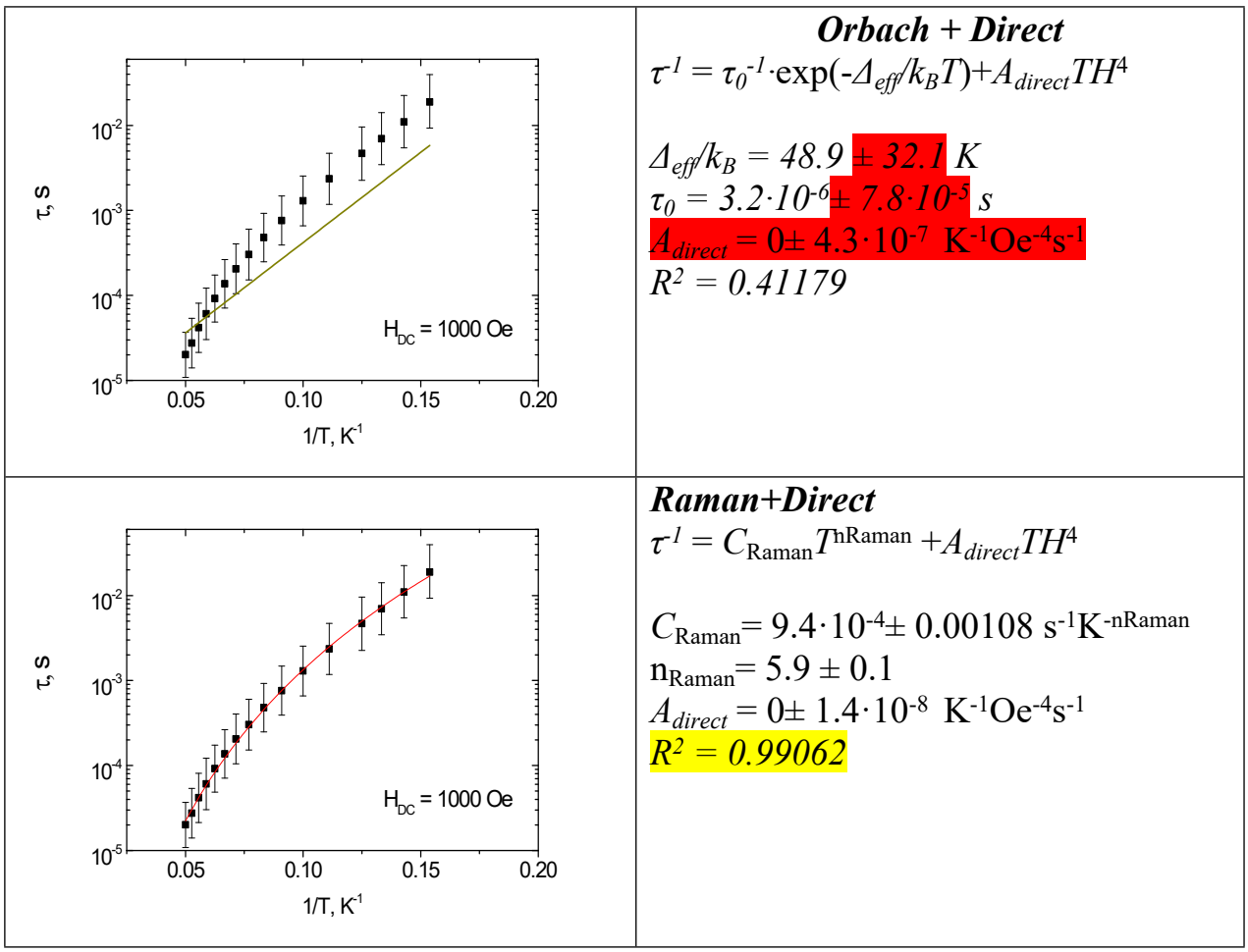
Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$
Δ_{eff}/k_B = 149 K (fixed)
$\tau_0 = 2.1 \cdot 10^{-8} \pm 2.4 \cdot 10^{-9}$ s
$C_{Raman} = 0.00682 \pm 3 \cdot 10^{-4}$ $s^{-1}K^{-nRaman}$
$n_{Raman} = 5$ (fixed)
$R^2 = 0.98039$



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$
$\Delta_{eff}/k_B = 47 \pm 0.5$ K (fixed)
$\tau_0 = 1.5 \cdot 10^{-5} \pm 1.3 \cdot 10^{-6}$ s
$C_{Raman} = 2.3 \cdot 10^{-6} \pm 1.4 \cdot 10^{-6}$ $s^{-1}K^{-nRaman}$
$n_{Raman} = 7.9 \pm 0.2$
$R^2 = 0.99962$



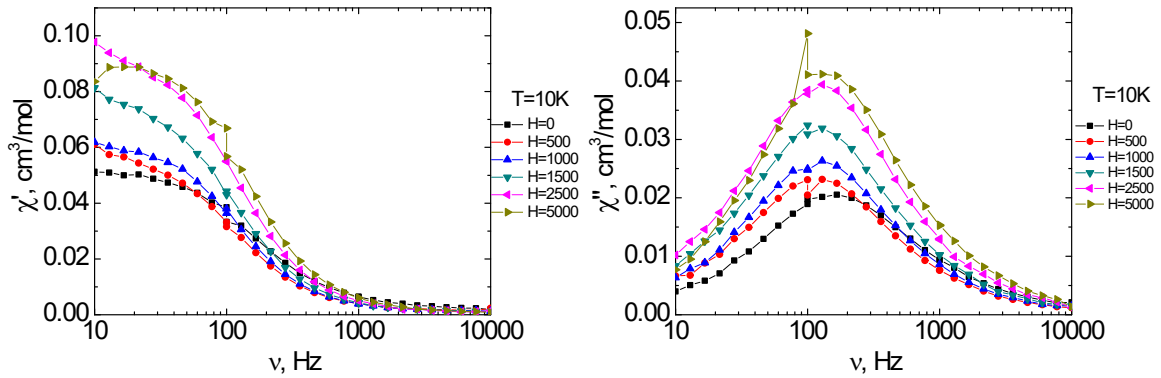


Fig. S31. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ'', right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.95}$** in various DC magnetic fields at 10 K. Solid lines are visual guides.

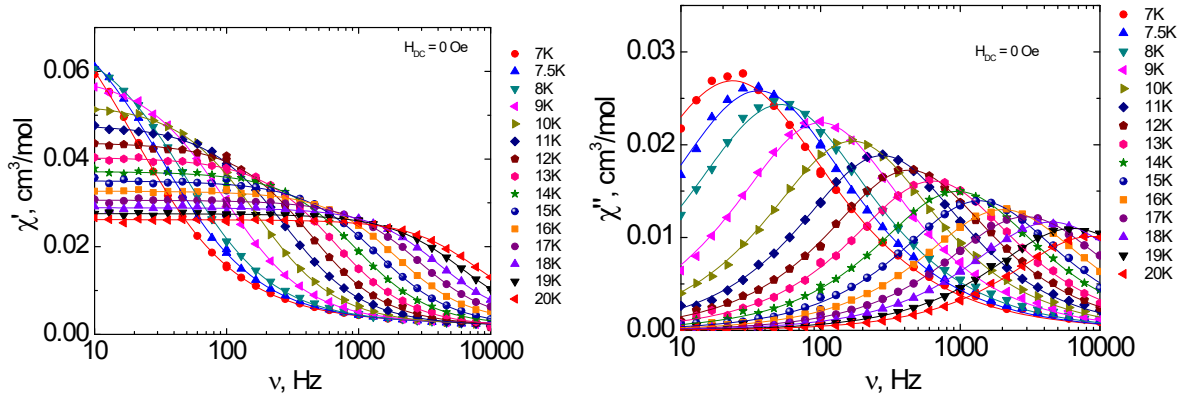


Fig. S32. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ", right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.95}$** in zero DC magnetic field. Solid lines show approximations of experimental data by the generalized Debye model.

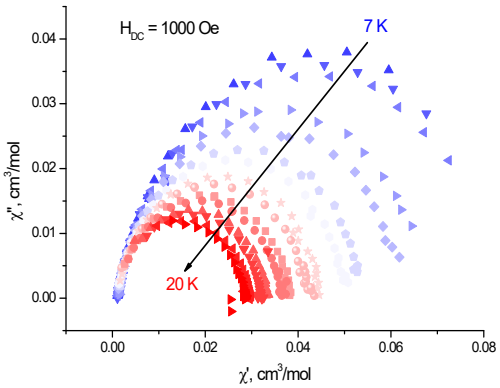
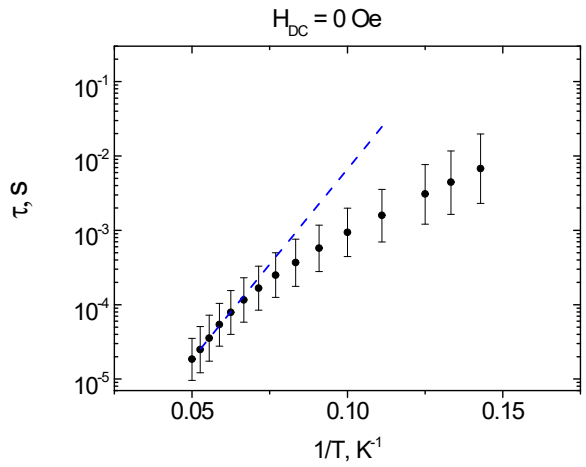


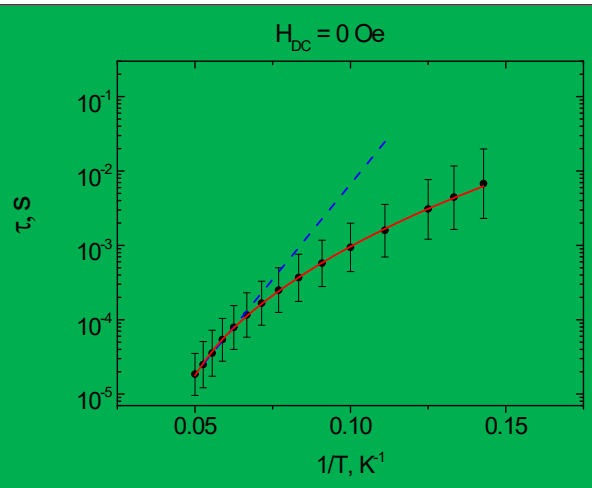
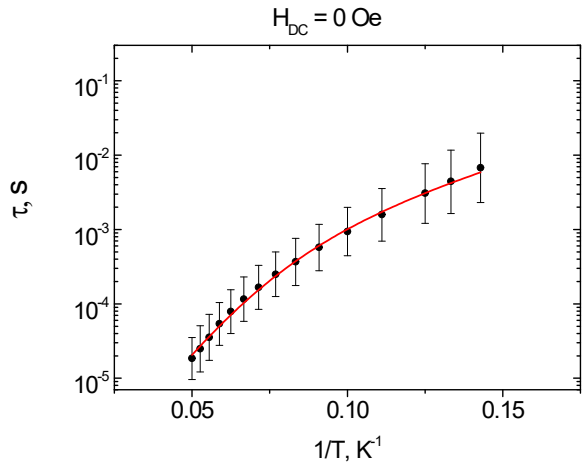
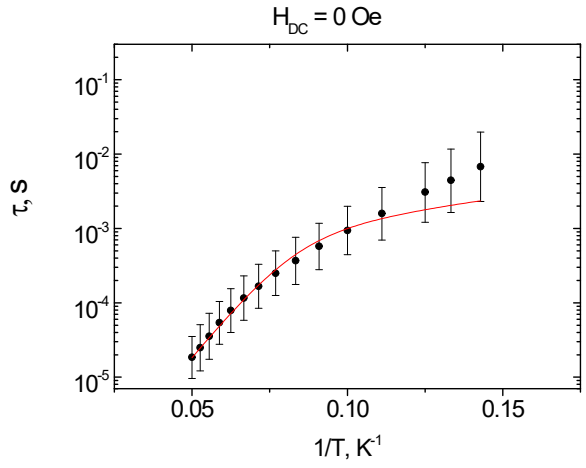
Fig. S33. Cole-Cole plots for **1_Y$_{0.95}$** in zero DC field.

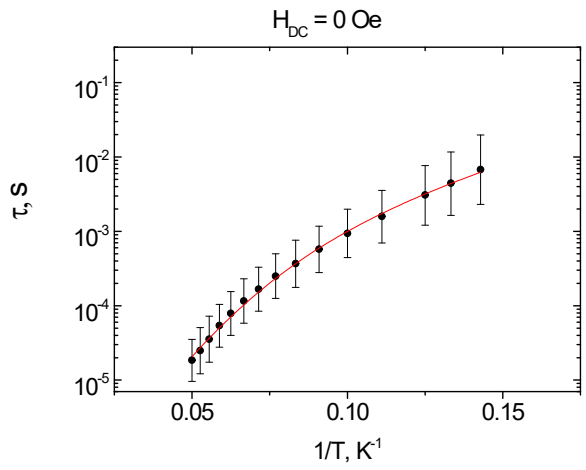
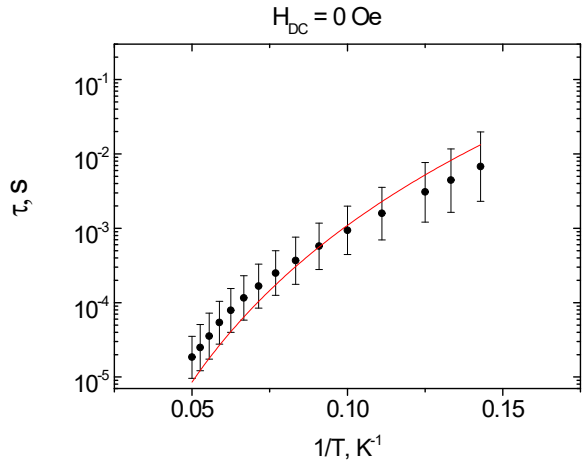
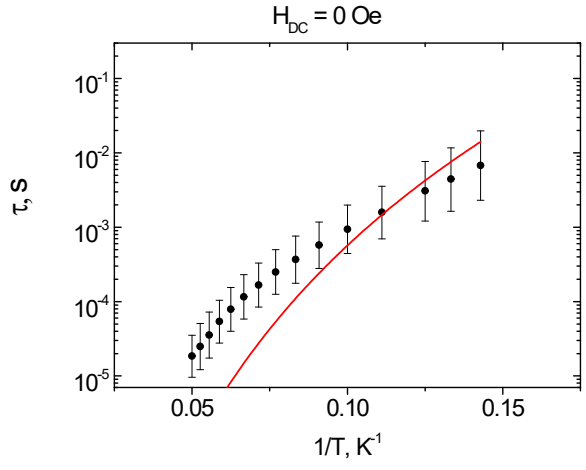
Table. S18. Distribution of parameter α at H_{DC} = 0 Oe for **1_Y$_{0.95}$**.

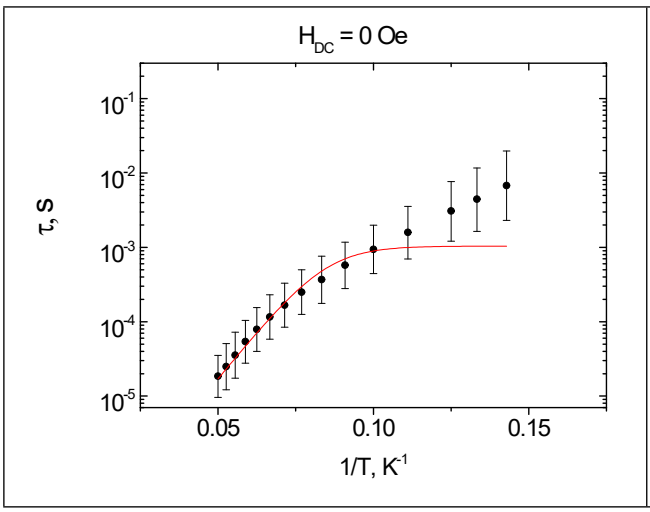
H_{DC} = 0 Oe	
T, K	α
7	0.21312
7.5	0.18364
8	0.16993
9	0.14056
10	0.12642
11	0.11871
12	0.11933
13	0.11528
14	0.10944
15	0.11298
16	0.10587
17	0.10355
18	0.11898
19	0.11671
20	0.09681

Table S19. Fitting of the τ *vs. 1/T* dependence for **1_Y$_{0.95}$** (H_{DC} = 0 Oe, T = 7-20 K). Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color.

Fitting line	**Mechanism(s)** Parameters
	**Orbach (18-20 K)** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT)$ $\tau_0 = 5.1 \cdot 10^{-8} \pm 6.2 \cdot 10^{-9}$ s $\Delta_{eff}/k_B = 118 \pm 2$ K $R^2 = 0.99932$

	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 265 K (fixed) $\tau_0 = 1.03 \cdot 10^{-10} \pm 1.3 \cdot 10^{-11}$ s $C_{Raman} = 0.0060 \pm 8 \cdot 10^{-4}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 5.24 \pm 0.05$ **R^2 = 0.99774**
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118 K (fixed) $\tau_0 = 1.16 \cdot 10^{-7} \pm 4.1 \cdot 10^{-8}$ s $C_{Raman} = 0.016 \pm 0.012$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 4.76 \pm 0.35$ R^2 = 0.98776
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118 K (fixed) $\tau_0 = 5.35 \cdot 10^{-8} \pm 4.5 \cdot 10^{-9}$ s $C_{Raman} = 8.7 \pm 1.1$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 2$ (fixed) R^2 = 0.93581

	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118 K (fixed) $\tau_0 = 1.5 \cdot 10^{-7} \pm 2 \cdot 10^{-8}$ s $C_{Raman} = 0.00953 \pm 3.96 \cdot 10^{-4}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 5$ (fixed) $R^2 = 0.98842$
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118 K (fixed) $\tau_0 = 1 \pm 1.6 \cdot 10^{7}$ s $C_{Raman} = 9.1 \cdot 10^{-5} \pm 2.0 \cdot 10^{-5}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 7$ (fixed) $R^2 = 0.59487$
	***Orbach+Raman*** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + C_{Raman}T^{nRaman}$ Δ_{eff}/k_B = 118 K (fixed) $\tau_0 = 1 \pm 2.5 \cdot 10^{8}$ s $C_{Raman} = 1.76 \cdot 10^{-6} \pm 1.09 \cdot 10^{-6}$ $s^{-1}K^{-nRaman}$ $n_{Raman} = 9$ (fixed) $R^2 = -0.19907$



Orbach+QTM

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT) + B$

$\Delta_{eff}/k_B = 118$ K

$\tau_0 = 4.7 \cdot 10^{-8} \pm 5 \cdot 10^{-9}$ s

$B = 961 \pm 200$ s^{-1}

$R^2 = 0.86268$

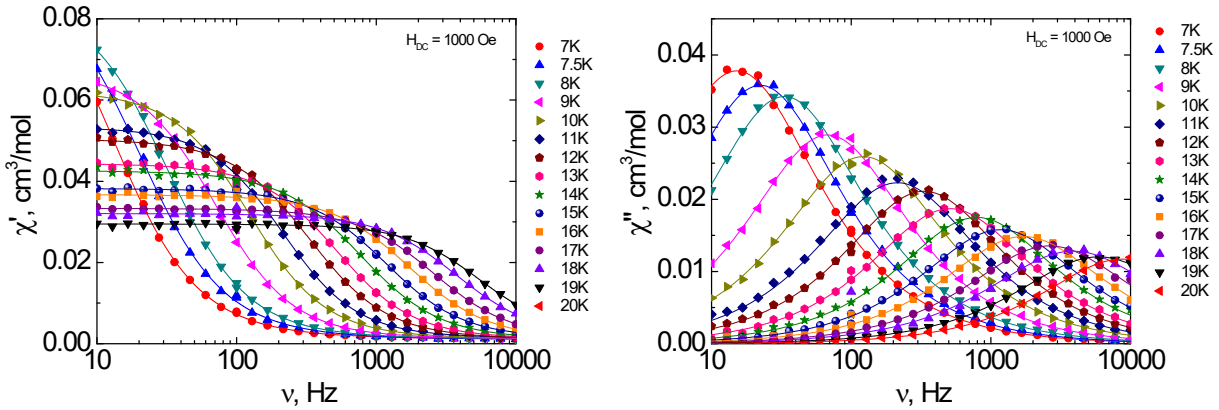


Fig. S34. Frequency dependencies of the in-phase (χ', left) and out-of-phase (χ'', right) components of dynamic magnetic susceptibility of complex **1_Y$_{0.95}$** in optimal DC magnetic field of 1000 Oe. Solid lines showapproximations of experimental databy the generalized Debye model.

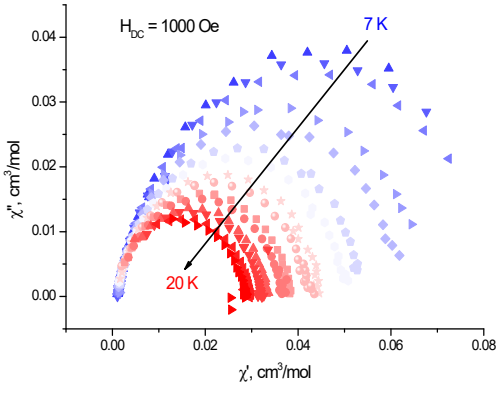
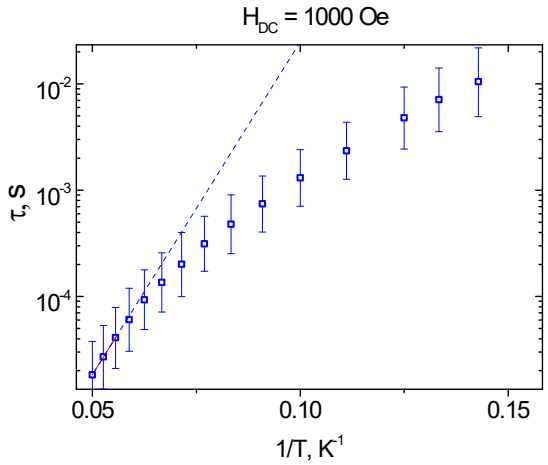


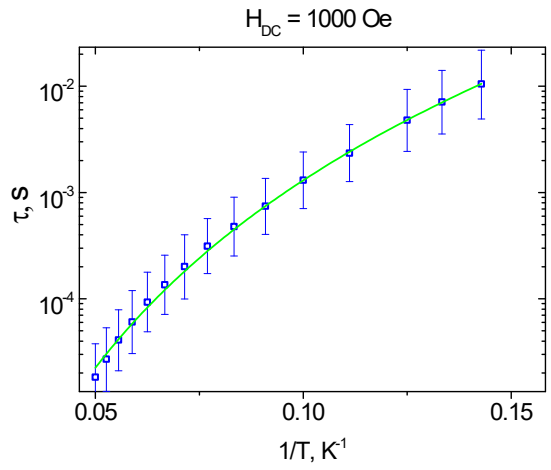
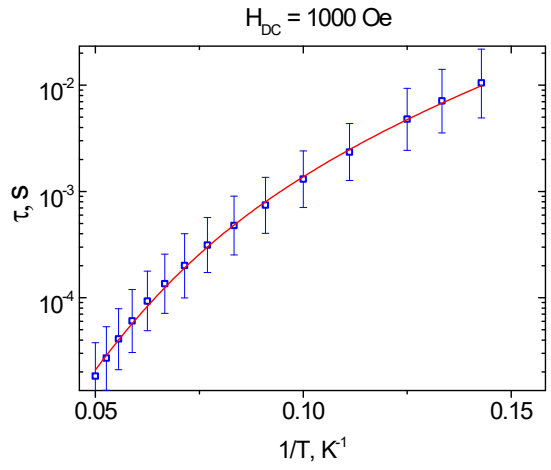
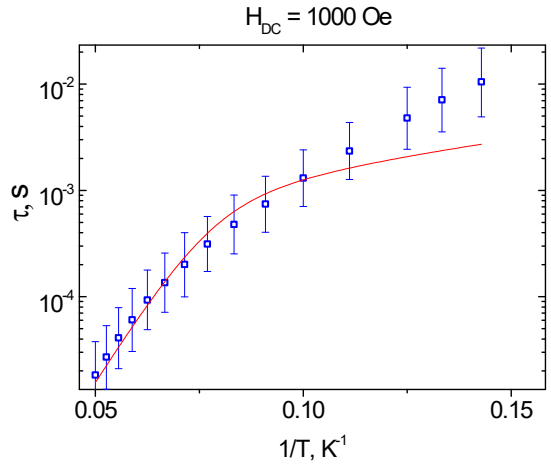
Fig. S35. Cole-Cole plots for **1_Y$_{0.95}$** in optimal DC magnetic field of 1000 Oe.

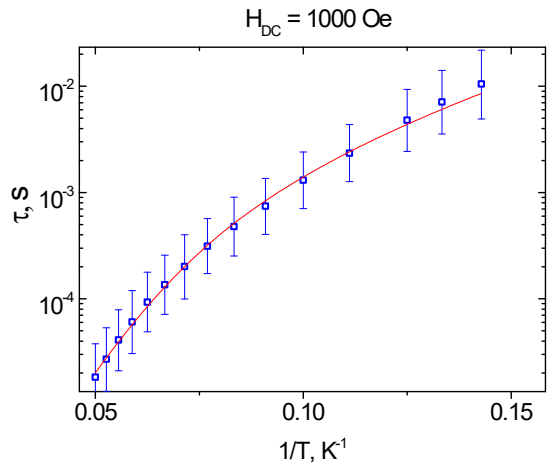
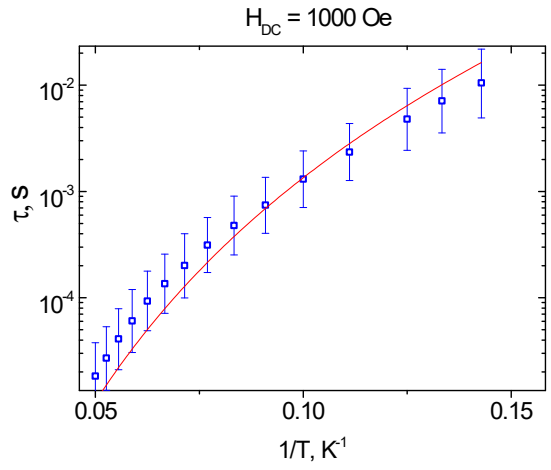
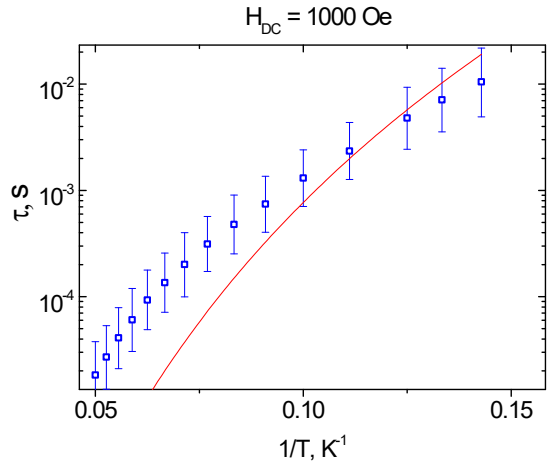
Table. S20. Distribution of parameter α at H_{DC} = 1000 Oe for **1_Y$_{0.95}$**.

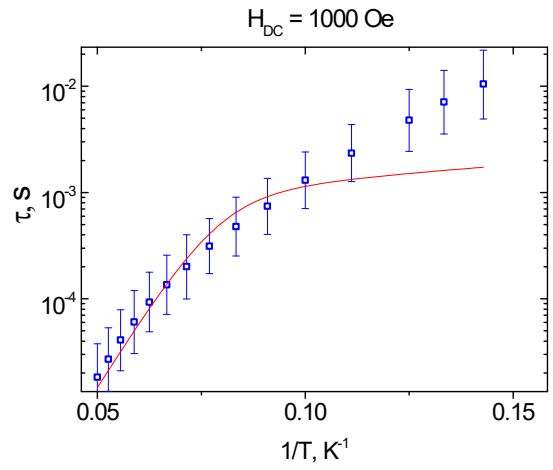
T, K	α
7	0.11733
7.5	0.1088
8	0.10544
9	0.09514
10	0.09361
11	0.08776
12	0.09907
13	0.09009
14	0.11128
15	0.10056
16	0.10416
17	0.10926
18	0.10007
19	0.10536
20	0.13067

Table S21. Fitting of the τ *vs. 1/T* dependence for **1_Y$_{0.95}$** (H_{DC} = 1000 Oe, T = 7-20 K). Meaningless parameters are specified in red color while yellow color emphasizes insufficient R^2 values. Reliable fits are specified in green color.

Fitting line	**Mechanism(s)** Parameters
	**Orbach (18-20 K)** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_BT)$ $\tau_0 = 1.36 \cdot 10^{-8} \pm 1.14 \cdot 10^{-9}$ s $\Delta_{eff}/k_B = 144 \pm 2$ K $R^2 = 0.9998$

	**Raman** $\tau^{-1} = C_{Raman} T^{nRaman}$ $C_{Raman} = 0.00106 \pm 7.4 \cdot 10^{-5} s^{-1} K^{-nRaman}$ $n_{Raman} = 5.86 \pm 0.036$ $R^2 = 0.99988$
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $\tau_0 = 4.7 \cdot 10^{-8} \pm 1.7 \cdot 10^{-8}$ s $\Delta_{eff}/k_B = 144$ K (fixed) $C_{Raman} = 0.00235 \pm 9.0 \cdot 10^{-4} s^{-1} K^{-nRaman}$ $n_{Raman} = 5.48 \pm 0.18$ $R^2 = 0.99266$
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $\tau_0 = 1.2 \cdot 10^{-8} \pm 2 \cdot 10^{-9}$ s $\Delta_{eff}/k_B = 144$ K (fixed) $C_{Raman} = 7.5 \pm 1.3 s^{-1} K^{-nRaman}$ $n_{Raman} = 2$ (fixed) $R^2 = 0.83035$

	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $\tau_0 = 2.7 \cdot 10^{-8} \pm 3 \cdot 10^{-9}$ s Δ_{eff}/k_B = 144 K(fixed) $C_{Raman} = 0.00694 \pm 2.4 \cdot 10^{-4} s^{-1} K^{-nRaman}$ n_{Raman} = 5 (fixed) R^2 = 0.98872
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $\tau_0 = 1 \pm 4.7 \cdot 10^{7}$ s Δ_{eff}/k_B = 144 K (fixed) $C_{Raman} = 7.4 \cdot 10^{-5} \pm 1.0 \cdot 10^{-5} s^{-1} K^{-nRaman}$ n_{Raman} = 7 (fixed) R^2 = 0.79565
	**Orbach+Raman** $\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{Raman} T^{nRaman}$ $\tau_0 = 1 \pm 1.2 \cdot 10^{9}$ s Δ_{eff}/k_B = 144 K (fixed) $C_{Raman} = 1.3 \cdot 10^{-6} \pm 4 \cdot 10^{-7} s^{-1} K^{-nRaman}$ n_{Raman} = 9 (fixed) R^2 = 0.20078



Orbach+Direct

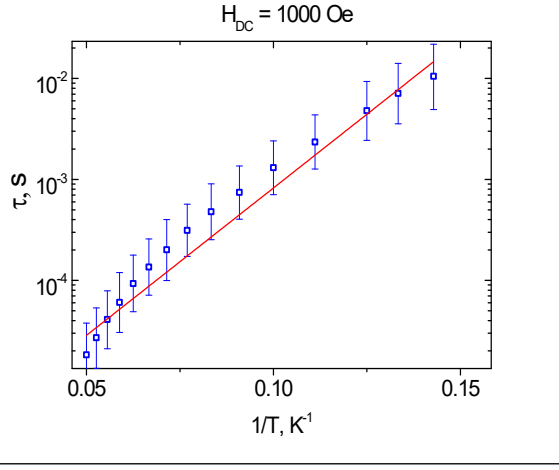
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + A_{direct} T H^4$

Δ_{eff}/k_B = 144 K (fixed)

$\tau_0 = 1.1 \cdot 10^{-8} \pm 2 \cdot 10^{-9}$ s

$A_{direct} = 8.3 \cdot 10^{-11} \pm 1.9 \cdot 10^{-11}$ $K^{-1}Oe^{-4}s^{-1}$

$R^2 = 0.76523$



Orbach+Direct

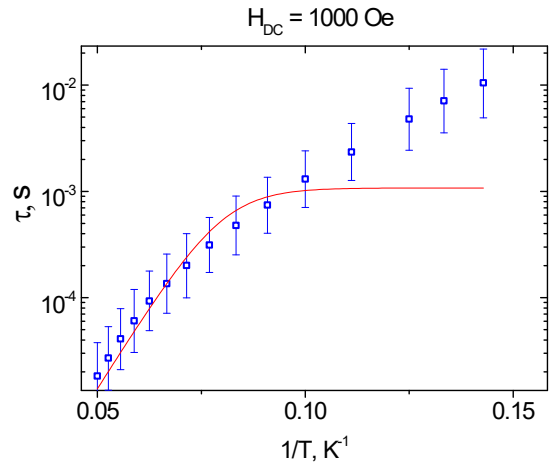
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + A_{direct} T H^4$

$\Delta_{eff}/k_B = 67.2 \pm 7.8$ K

$\tau_0 = 1 \cdot 10^{-6} \pm 5.7 \cdot 10^{-6}$ s

$A_{direct} = 0 \pm 1 \cdot 10^{-7}$ $K^{-1}Oe^{-4}s^{-1}$

$R^2 = 0.80697$



Orbach+QTM

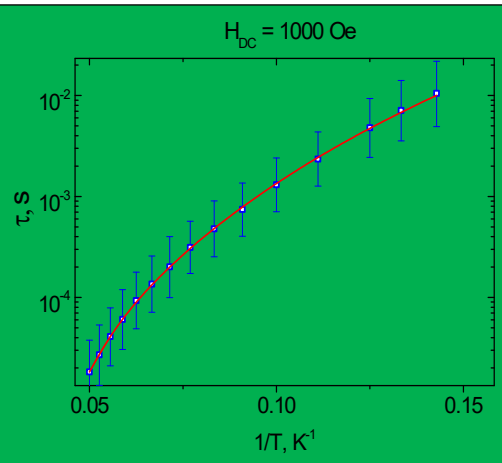
$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + B$

$\Delta_{eff}/k_B = 144$ K (fixed)

$\tau_0 = 1.05 \cdot 10^{-8} \pm 2.3 \cdot 10^{-9}$ s

$B = 928 \pm 259$ s^{-1}

$R^2 = 0.70574$



Orbach+Raman

$\tau^{-1} = \tau_0^{-1} \cdot \exp(-\Delta_{eff}/k_B T) + C_{\mathrm{Raman}} T^{\mathrm{nRaman}}$

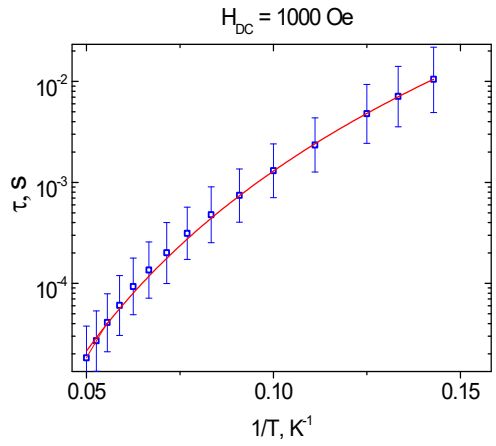
Δ_{eff}/k_B = 265 K (fixed)

$\tau_0 = 1.0 \cdot 10^{-10} \pm 1 \cdot 10^{-11}$ s

$C_{\mathrm{Raman}} = 0.0017 \pm 2 \cdot 10^{-4}$ $s^{-1}K^{-\mathrm{nRaman}}$

$n_{\mathrm{Raman}} = 5.64 \pm 0.04$

R^2 = 0.99857



Raman+Direct

$\tau^{-1} = C_{\mathrm{Raman}} T^{\mathrm{nRaman}} + A_{direct} T H^4$

$C_{\mathrm{Raman}} = 9.0 \cdot 10^{-4} \pm 2.5 \cdot 10^{-4}$ $s^{-1}K^{-\mathrm{nRaman}}$

$n_{\mathrm{Raman}} = 5.93 \pm 0.13$

$A_{direct} = 3.32 \cdot 10^{-13} \pm 5.3 \cdot 10^{-13}$ $K^{-1}Oe^{-4}s^{-1}$

$R^2 = 0.99987$