Supplementary information

Effect of magnetic fields on the methyl rotation in a paramagnetic cobalt(II) complex. Quasielastic neutron scattering studies

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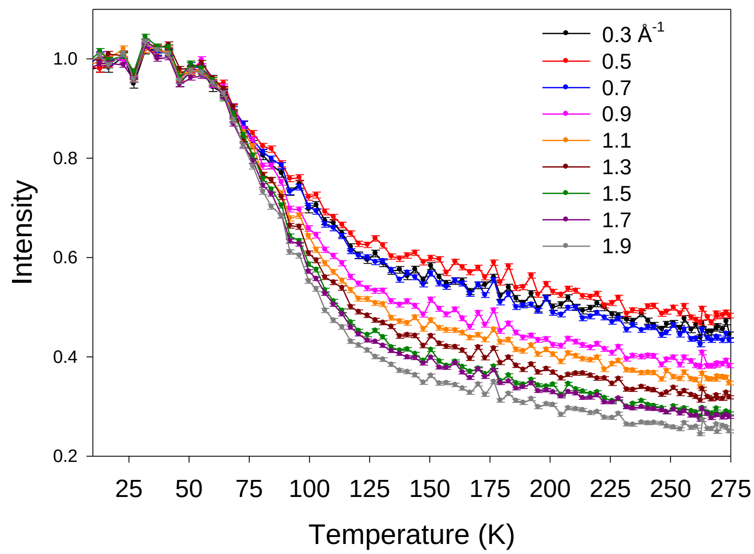

Fig. S1 Fixed window elastic scattering neutron intensity scan at variable $|Q|$.

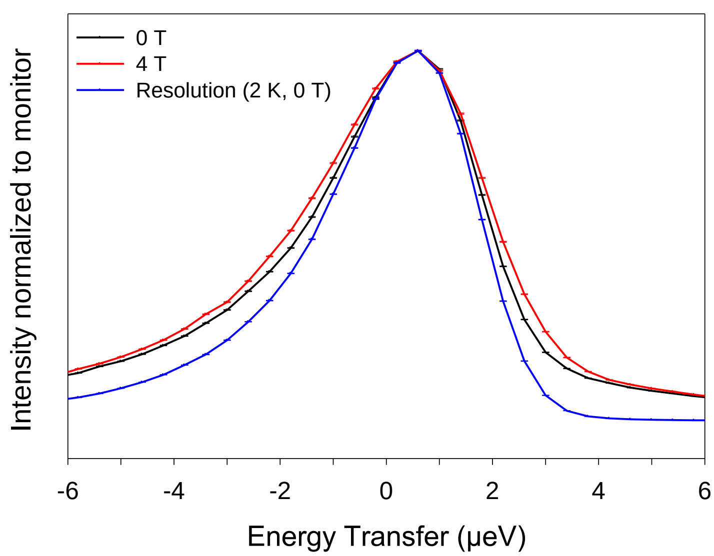

Fig. S2 Comparison of resolution function at 2 K with QENS data at 100 K for 0 and 4 T. The data presented here is zoomed in around the elastic line to emphasize the difference in the broadening.

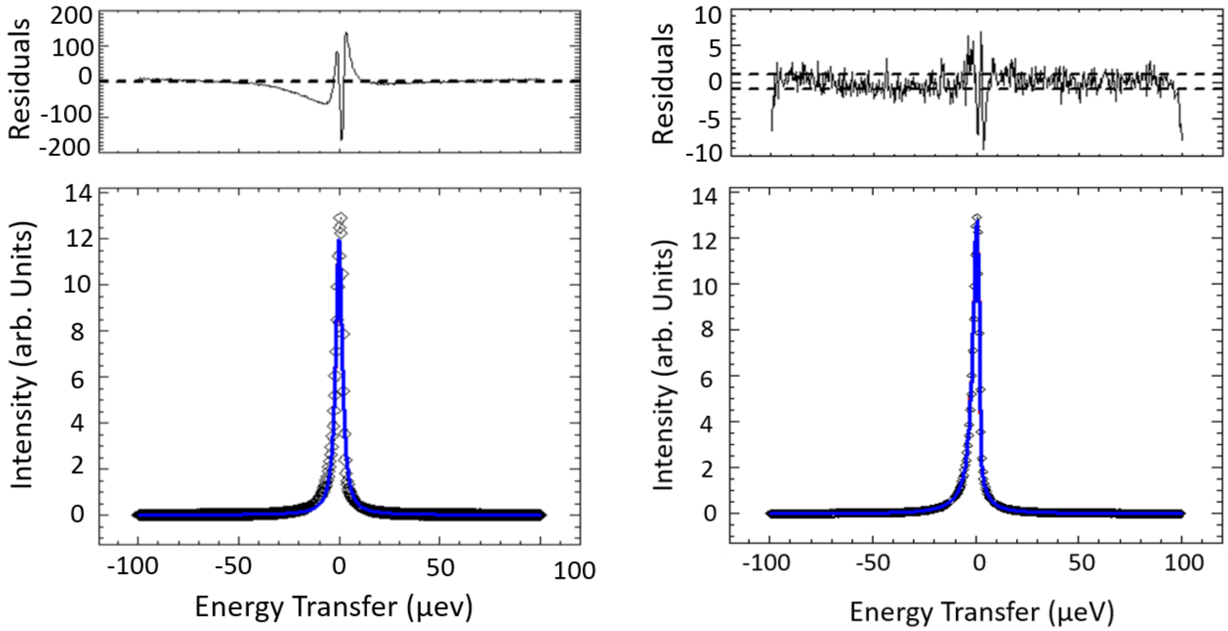

Fig. S3 (Left) The rejected Lorentzian model (blue line) to fit a representative data set at 100 K and 0 T. The difference between the data and the model is shown on the top panel. (Right) The use of the Cole-Cole equation (blue line) to fit the aforementioned data set. The parameters from these fittings are given in Table S1.

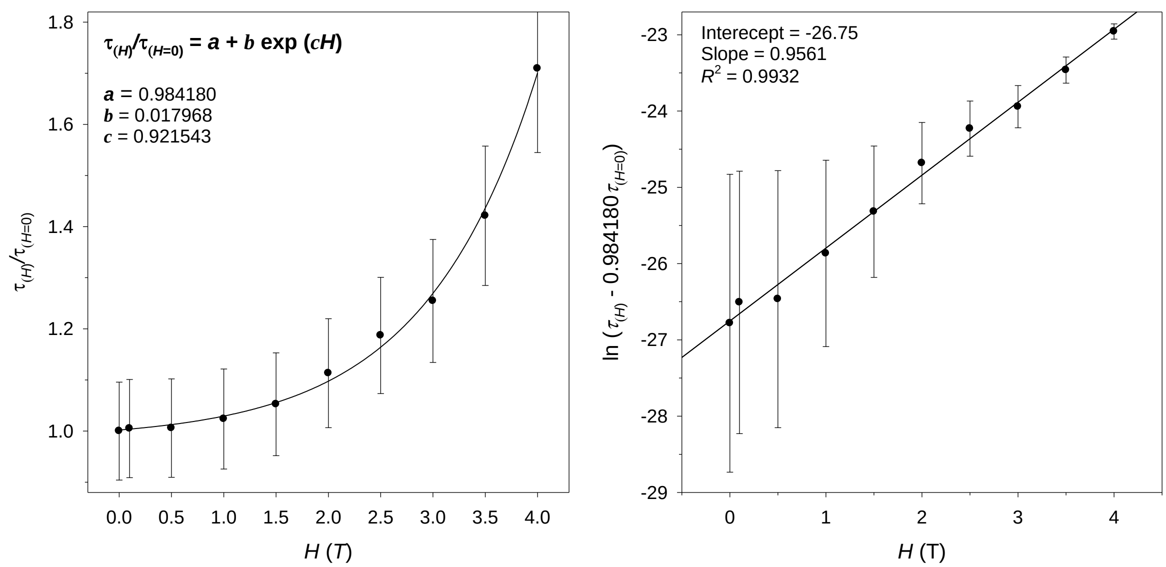

Fig. S4 (Left) Plot of $\tau_{(H)}/\tau_{(H=0)}$ vs. H at 100 K. a, b and c are fitting constants. (Right) Plot of ln ($\tau_{(H)} - 0.984180\tau_{(H=0)}$) vs H. The number a = 0.984180 is from the fitting of Fig. S4-Left.

Table S1 Comparison of the broadening and elastic parameters of the QENS peak at different fields. The error in the E_0 and elastic parameters are in parentheses.

80 K, summed over all *Q*				
Field (T)	**E_0, μeV[a]**	**α**	**Elastic scattering fraction, *x***	**τ (s)**
0.0	1.19(6)	7.8(4) x 10^{-2}	0.632(4)	5.6(3) x 10^{-10}
4.0	0.323(17)	8.3(3) x 10^{-2}	0(0)	2.04(10) x 10^{-9}
90 K, summed over all *Q*				
Field (T)	**E_0, μeV[a]**	**α**	**Elastic scattering fraction, *x***	**τ (s)**
0.0	2.43(12)	7.6(3) x 10^{-2}	0.577(2)	2.71(13) x 10^{-10}
4.0	1.14(6)	1.7(3) x 10^{-1}	0.332(5)	5.8(3) x 10^{-10}
100 K, summed over all *Q*				
Field (T)	**E_0, μeV[a]**	**α**	**Elastic scattering fraction, *x***	**τ (s)**
0.0	4.46(22)	9.2(2) x 10^{-2}	0.537(1)	1.48(7) x 10^{-10}
0.1	4.43(22)	8.9(2) x 10^{-2}	0.541(1)	1.49(8) x 10^{-10}
0.5	4.43(22)	8.4(2) x 10^{-2}	0.543(1)	1.49(8) x 10^{-10}
1.0	4.35(22)	8.8(2) x 10^{-2}	0.538(1)	1.51(8) x 10^{-10}
1.5	4.23(21)	8.8(2) x 10^{-2}	0.537(1)	1.55(8) x 10^{-10}
2.0	4.00(20)	1.1(2) x 10^{-1}	0.515(1)	1.64(8) x 10^{-10}
2.5	3.75(19)	1.4(2) x 10^{-1}	0.490(2)	1.75(9) x 10^{-10}
3.0	3.55(18)	1.4(2) x 10^{-1}	0.484(2)	1.85(9) x 10^{-10}
3.5	3.13(16)	1.7(2) x 10^{-1}	0.446(2)	2.10(11) x 10^{-10}
4.0	2.61(13)	2.0(3) x 10^{-1}	0.396(2)	2.53(12) x 10^{-10}

[a] Total uncertainties σ_{total} in E_0 are given in Table S1 here:[8] $\sigma_{total}^2 = \sigma_{ran}^2 + \sigma_{sys}^2$. Random uncertainty σ_{ran} for each E_0 value is obtained from the fitting of the QENS data using Eq. 1. Systematic uncertainty σ_{sys} in E_0 from the QENS studies here is estimated to be 5% of E_0.

Analysis of the elastic scattering fraction, *x*, between 0 and 4 T in Table S1 shows that the elastic fraction (*x*) is reduced with magnetic field at all three temperatures, suggesting that more methyl groups activated by the application of the magnetic field. Yet, the methyl rotation becomes slower with the field. At this time, we cannot explain this trend. Since τ, x and α are co-dependent parameters, we have investigated the fitting of the QENS data with the elastic parameter (*x*) fixed at the value at 0 T for the three temperatures. Representative results of this fitting for the data at 80 K are given in Fig. S5. The chi-squared values χ^2 for the fitting at all three temperatures are given in Table S2 below.

The fitting with fixed *x* is comparatively worse. Not only are chi-squared values χ^2 much higher in the fixed *x*, but also there is a large systematic deviation between the data and the model, which is evident in the difference plots (Fig. S5-Top) demonstrating the inadequacy of the fits with the elastic fraction *x* fixed at 0 T. Therefore we have used an unfixed *x* for the QENS fittings in Table S1.

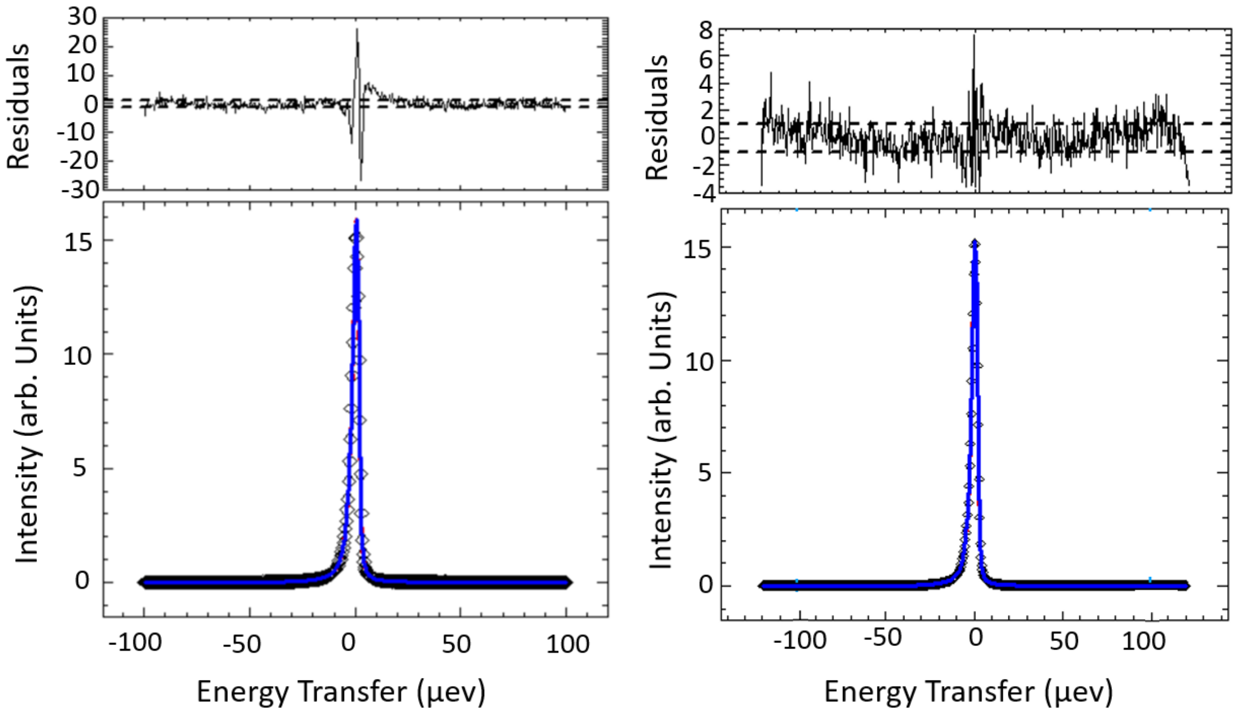

Fig. S5 (Left) Fitting (blue line) of the QENS data at 80 K with *x* fixed at the values at 0 T using the Cole-Cole equation. The difference between the data and the model is shown on the top. (Right) Fitting (blue line) of the same data with *x* unfixed using the Cole-Cole equation.

Table S2 Chi-squared χ^2 values at 4 T for fixed and unfixed elastic parameter (*x*) fittings

Temperature (K)	Fixed *x*	Unfixed *x*
80	10.554	1.632
90	32.186	3.332
100	17.612	5.124

Table S3 Average spin density $\rho_{s\text{-average}}$ per atom in **1**.

Atom	$\boldsymbol{\rho_{s\text{-average}}}$
H6	-1.28 x 10^{-5}
H3-5 or H7-9	7.16 x 10^{-5}
H1-2	9.00 x 10^{-4}
C6	3.40 x 10^{-3}
C2-3	-9.34 x 10^{-4}
C1,5	2.10 x 10^{-3}
O2-3	2.70 x 10^{-2}
O1	1.91 x 10^{-1}
Co1	2.81

References for ESI

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