# Muon LCR measurements for organic magnets based on [Pd(dmit)₂] metal-complex molecules†

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Magnetic ground states of quasi two dimensional (Q2D) triangular Heisenberg antiferromagnetic (AF) systems are of great interest. Magnetic frustration arising from the triangular exchange network suppresses the AF order. This kind of quantum-spin states without either long-range magnetic order or lattice symmetry breaking is named quantum spin liquid (QSL) state. Theoretically, this QSL state has been studied extensively and many classes of theoretical models such as $Z_2$ spin liquid, spinon fermi surface (SFS) and spin-bose metal are proposed. Although experimentalist have sought real model materials with QSL state for quite some time, only a few candidate materials are known to this date.

A series of organic salts, $(Cation)[\text{Pd}(\text{dmit})_2]_2$ (dmit=1,3-dithiole-2- thione-4,5-dithiolate) has triangular exchange network of S = 1/2 unit of molecular dimers[1]. The strength of the spin frustration can be controlled by the choice of cation and most of materials belonging to this family undergo AF states. In such triangular magnets, geometrical frustration is though to play an important role on the magnetic state as the AF transition temperatures is found to increase proportionally to the deviation from the regular triangular exchange networks. It is thought that EtMe₃Sb[Pd(dmit)₂]₂[2] do not show any AF order due to strong spin frustrations. These materials have almost regular triangular exchange networks with exchange interaction $J$ of the order of 200 to 300 K.

Recently, we have performed longitudinal field (LF) $\mu$SR measurements on EtMe₃Sb[Pd(dmit)₂]₂ a QSL candidate. Preliminary analysis suggest that field dependence of muon relaxation rate, $\lambda$ is proportional to $1/\sqrt{B}$ behaviour in a field range of $1 \leq B_{ext} \leq 1000$ Gauss at low-temperatures. Such a behaviour is expected from spins diffusing along a one-dimensional direction. In an ideal 1D spin system model, the field dependence of $\lambda$ is approximately described as follows:

$$\lambda(\omega) = \frac{A^2}{4} \frac{1}{\sqrt{2D_{\parallel}\omega}} \qquad (1)$$

where $D_{\parallel}$ is the diffusion rate of spinon, $\omega = \gamma_e B$ is the Larmor frequency and $A$ is the scalar hyperfine coupling constant[3]. In order to determine $D_{\parallel}$ and its temperature dependence quantitatively, a Muon level-crossing resonance measurements (LCR) of the Pd(dmit)₂ molecule was made. For this purpose, we used (EtMe₃Sb)₂[Pd(dmit)₂], a non-magnetic material, instead of the neutral Pd(dmit)₂ molecule due to chemical stability issues.
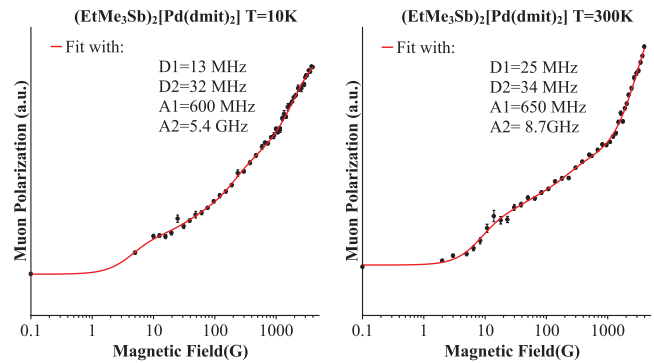


Fig. 1. Longitudinal polarization for the muon radical in (EtMe₃Sb)₂[Pd(dmit)₂] at 10K and 300K together with fitted repolarization curves..

Results of the longitudinal field dependence in (EtMe₃Sb)₂[Pd(dmit)₂] at port-2 of the RIKEN-RAL Muon facility are shown in Fig. 1. The Muon repolarization curve show no resonance in fields up to 0.4 Tesla and show an incredibly broad distribution of hyperfine couplings at 10K. Preliminary DFT calculation with muon radicals at the sulphur ends of the $[\text{Pd}(\text{dmit})_2]^{-2}$ molecule predicts anisotropic hyperfine parameters (A, D1, D2)=(360, 11, 5) MHz and a resonance around 1T. Fit of the curves require at least a four-term equation with hyperfine values shown within the graph. The deduced values appear to be at least twice those found from the DFT calculation which support the hypothesis of the Muonium formation at the sulphur ends of the $[\text{Pd}(\text{dmit})_2]^{-2}$ molecule. However another large hyperfine term with GHz values must be added to fit the repolarization curve indicating at least one more predominant Muonium site with a higher field resonance.

Further LCR measurements, in magnetic fields up to 5 Tesla in the Hi-Fi magnet at the ISIS muon facility have recently been awarded and will complete this study to understand the Muonium sites on a Pd(dmit)₂ molecule and the hyperfine coupling constant associated with such sites.

## References
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