Study of frustrated antiferromagnetic states by μSR

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A series of organic salts, $(Cation)[Pd(dmit)_2]_2$ (dmit=1,3-dithiole-2-thione-4,5-dithiolate) has a triangular exchange network of S = 1/2 units of molecular dimers[1]. The strength of the spin frustration can be controlled by the choice of the cation. In such triangular magnets, geometrical frustration is thought to play an important role in determining the magnetic state, as the antiferromagnetic (AF) transition temperature is found to increase proportionally to the deviation from the almost regular triangular exchange networks. It is thought that $EtMe_3Sb[Pd(dmit)_2]_2$[2] does not show any AF order owing to strong spin frustrations. The exchange interaction J in these materials is on the order of 200 to 300 K.

Recent longitudinal field (LF) μSR measurements were performed on $EtMe_3Sb[Pd(dmit)_2]_2$, a quantum spin liquid (QSL) candidate. Preliminary analysis suggests that the field dependence of the muon relaxation rate, λ, is proportional to $1/\sqrt{B}$ in a field range of $1 \leq B_{ext} \leq 1000$ G at low-temperatures. Such behaviour is expected from one-dimensional (1D) spin diffusion, a surprising result considering the quasi two dimensional nature of the electronic structure of $Pd(dmit)_2$-based compounds.

To study how geometrical frustration affects the nature of the spin fluctuations, we have performed μSR measurements on $Et_2Me_2P[Pd(dmit)_2]_2$, a Mott insulator with an AF transition of 14 K. Analysis in the paramagnetic temperature region of the μSR spectra was performed using the following formula:

$$A(t) = A_0 \exp(-\lambda t) \times G_{KT}(\Delta, H_{LF}, t)$$

where $G_{KT}(\Delta, H_{LF}, t)$ is the Kubo-Toyabe function, which is due to nuclear dipoles, and λ is the muon depolarization rate, which describes the degree of electron spin fluctuation. Δ values were determined with the zero field (ZF) data and were assumed to be LF independent.

Figure 1 shows that the LF dependence of λ at 100 K in $Et_2Me_2P[Pd(dmit)_2]_2$ is a combination of two different spin fluctuation motions. At low fields, λ follows a power law LF dependence with an exponent of -0.86, a value close to the -1/2 value expected from a spinon 1D spin diffusion motion, whereas at higher magnetic fields a logarithmic dependence of λ emerges, indicative of a purely 2D spin diffusion or 1D ballistic spin motion[3,4]. The inset of Fig. 1 shows that the temperature dependence of λ at ZF, which is dominated by the 1D spin diffusion motion, increases with decreasing temperature in contrast to the temperature dependence of λ at 100 G, which peaks at 100 K and is dominated by 2D spin diffusion or 1D ballistic spin motion. This 100 K maximum of λ at 100 G is interesting as it correlates with the thermoelectric power maximum and the onset of the anomalous dielectric constant[5] of the same compound.

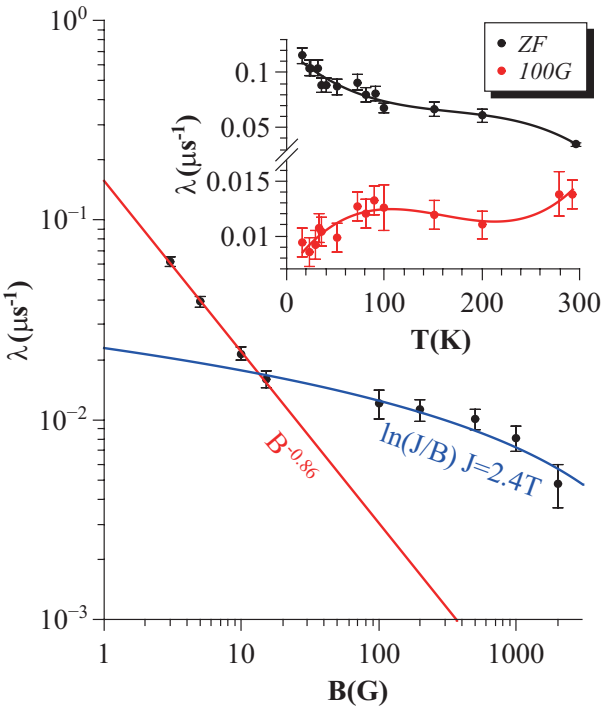

Fig. 1. Dependence of the longitudinal relaxation rate λ on the applied longitudinal field at 100 K in $Et_2Me_2P[Pd(dmit)_2]_2$. (Inset) Temperature dependence of the relaxation rate λ of $Et_2Me_2P[Pd(dmit)_2]_2$ at a zero field and a 100 G longitudinal field.

LF μSR measurements in $Et_2Me_2P[Pd(dmit)_2]_2$ have shown that decreasing geometrical frustration on the spin-liquid level not only increases the AF transition temperature but changes the nature of the spin fluctuations from the purely 1D diffusion motion of a spinon. Further, LF μSR measurements are required to properly quantify the ratio between the two different spin motions, their relation to charge transport, and the degree of geometrical frustration.

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References

1) K. Kanoda and R. Kato, Annu. Rev. Condens. Matter Phys. **2**, 167 (2011).
2) M. Yamashita et al., Science, **328**, 1246 (2010).
3) F. L. Pratt et al., Phys. Rev. Lett. **96**, 247203 (2006).
4) F. L. Pratt et al., Physica B **404**, 585589 (2009).
5) M. Abdel-Jawad et al., Phys. Rev. B **88**, 075139 (2013).