Muon spin relaxation study on the new organic spin liquid material \(\lambda-(\text{STF})_2\text{GaCl}_4\)

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Research on quantum spin liquid (QSL) states in frustrated quantum magnetism is a critical issue in the field of condensed matter physics. In recent research, some candidate QSL materials have been found in molecular-based organic systems, such as \((\text{ET})_2\text{Cu}_2(\text{CN})_3\)3 and \(\text{EtMe}_3\text{Sb}[\text{Pd(dmit)}_2]\)2.2 The spin systems in these salts have been well described as an antiferromagnetic (AF) spin system with a nearly equilateral regular triangular lattice. Recently, we found a new candidate QSL material, \(\lambda-(\text{STF})_2\text{GaCl}_4\), which is related to the organic superconductor \(\lambda-(\text{BETS})_2\text{GaCl}_4\). As shown in Fig. 1, down to 2 K, the static susceptibility of \(\lambda-(\text{STF})_2\text{GaCl}_4\) is very well described by the AF Heisenberg model with a regular triangular lattice, which is the typical behavior of the QSL system.3 In contrast to the good agreement of the susceptibility behavior, the calculated geometry of the spin interaction in \(\lambda-(\text{STF})_2\text{GaCl}_4\) is quite distorted from the regular triangle. Therefore, it can be the key material to investigate the stabilization mechanism of the QSL state.

We succeeded in synthesizing a high-quality sample of \(\lambda-(\text{STF})_2\text{GaCl}_4\) with the total mass \(\sim 75 \text{ mg}\) and conducted ZF-\(\mu\)SR measurement down to 0.3 K. Figure 2 shows the \(\mu\)SR time spectra measured at 0.3 K in ZF and under an LF of 100 G. The ZF-\(\mu\)SR time spectrum is well described by the simple exponential function. Since the \(\mu\)SR time spectrum is well decoupled by applying LF of 100 G, there is no static internal field due to AF long-range ordering. On the other hand, we can see a slow relaxation behavior even at 0.3 K under an LF of 100 G. This implies that there are some effects of fluctuating internal fields originating from surrounding electronic spins, and the sample does not show the long-range AF ordering although it has a strong AF interaction \(J/k_B \sim -165 \text{ K}\), at least down to 0.3 K.

This behavior is similar to that observed in the \(\mu\)SR measurement for the first organic QSL salt, \(\kappa-(\text{ET})_2\text{Cu}_2(\text{CN})_3\)4. Additionally, the authors of Ref. 4 found that the LF-\(\mu\)SR time spectrum is described by a two-component exponential function and pointed out that the system undergoes phase separation into paramagnetic islands and a singlet phase at a low temperature. Therefore, we are now planning an additional experiment for \(\lambda-(\text{STF})_2\text{GaCl}_4\) to observe the spin behavior in the range between ZF and an LF of 100 G. With the progress of this research, the microscopic spin dynamics in the QSL state and the stabilization mechanism of the QSL state in a distorted lattice system can probably be clarified.

References