Muon spin rotation study on the new organic spin liquid candidate λ-$(STF)_2GaCl_4$

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In recent research, some candidate quantum spin liquid (QSL) materials have been found in molecular conductors such as κ-$(ET)_2Cu_2(CN)_3$[1)] and $EtMe_3Sb[Pd(dmit)_2]_2$,[2)] which have antiferromagnetic (AF) spin systems with a nearly regular triangular lattice. Recently, we found a new candidate QSL material, λ-$(STF)_2GaCl_4$, which can be located between the organic superconductor λ-$(BETS)_2GaCl_4$ and the antiferromagnet λ-$(ET)_2GaCl_4$ in the temperature-pressure phase diagram shown in Fig. 1. The AF phase in the ET salt is suppressed by a small pressure, and the STF salt shows neither the AF nor the superconducting transition down to 1.6 K. The STF salt shows a gradual peak structure in the temperature dependence of the static susceptibility, which is well described by the AF Heisenberg model with a regular triangular lattice.[3)] In contrast to the good agreement of the susceptibility behavior, the calculated geometry of the spin interaction in λ-$(STF)_2GaCl_4$ is quite distorted from a regular triangle. Therefore, it can be a key material to investigate the stabilization mechanism of the QSL state. Hence we conducted a μSR study to investigate the local spin dynamics of λ-$(STF)_2GaCl_4$.

We succeeded in synthesizing a high-quality sample of λ-$(STF)_2GaCl_4$, and in the 2017 beamtime, we conducted the first μSR measurement. The μSR time spectrum at 0.3 K started to decouple on applying a longitudinal field (LF) of 100 G, indicating that there is no AF long-range ordering. However, by comparing the time spectrum with that in an LF of 1000 G, for which the complete decoupling behavior was observed, we found that a slow relaxation remains, indicating that there are some effects of fluctuating internal fields. This behavior is similar to that observed in the μSR measurement of κ-$(ET)_2Cu_2(CN)_3$.[4)]

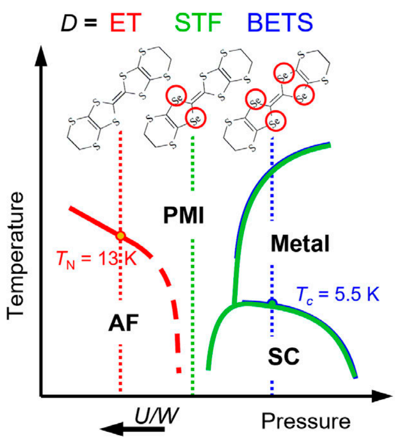

Fig. 1. Schematic temperature-pressure phase diagram of λ-D_2GaCl_4 (D = ET, STF, BETS).[3)]

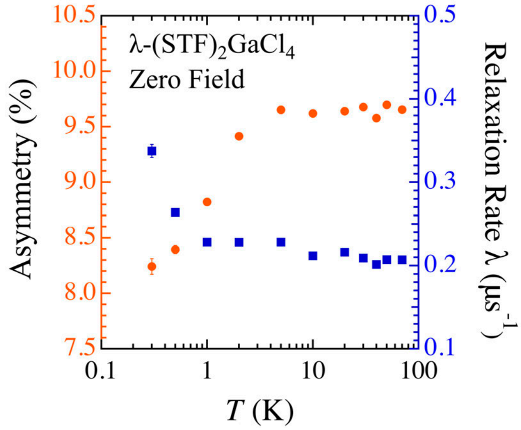

Fig. 2. Temperature dependence of the μSR relaxation rate and total asymmetry under zero field in λ-$(STF)_2GaCl_4$.

In the beamtime in July 2019, we conducted high-statistics μSR measurement and improved the experimental setup. The total asymmetry was improved in comparison with the previous experiment in 2017, from 6% to 8.3% at 0.3 K. A resonable asymmetry of about 10% is frequently observed in molecular conductors due to the formation of muonium. The time spectra can be fitted by a single exponential function. Figure 2 shows the temperature dependence of the total asymmetry and relaxation rate λ. The total asymmetry at a high temperature was 9.3% and started to decrease below 5 K. This decrease was not observed in the previous measurement due to unsatisfactory experimental conditions. In conjunction, we observed the increase of λ. We now suspect that these give evidence for the gradual growth of the local staggered moments in the short-range AF ordered region. The lack of the transition to the bulk AF state with the growth of the short-range order is considered to be due to static spin heterogeneity resulting from the disorder or due to the effect of the spin frustration. As the next step, it is important to measure the spin dynamics toward the lower-temperature region to elucidate whether the long-range AF order or the quantum critical point exists. Now, we have submitted a proposal for measurement using a $^3He/^4He$ dilution refrigerator. With the progress of this research, the microscopic spin dynamics in the QSL state and the stabilization mechanism of the QSL state in a disordered system can probably be clarified.

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