Zero-field μSR on single-crystal organic superconductor λ-$(BETS)_2GaCl_4$

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Unconventional superconductivity is often realized in the vicinity of a magnetic ordered state, as observed in ruthenates, heavy fermions, cuprates, organic superconductors, and Fe-based superconductors.[1)] In the organic superconductor family κ-$(ET)_2X$ with the cation molecules ET = $(CH_2)_2S_4C_6S_4(CH_2)_2$ and anion molecules X, the superconducting state emerges on applying pressure to the long-range-ordered antiferromagnetic insulator at a low temperature below about 10 K, while at a high-temperature region, a crossover occurs from a metallic to a paramagnetic insulating state.[2)] Therefore, it is suspected that the metallic-state spin correlations govern the superconductivity.

Powell and McKenzie[3)] proposed that for the whole family of organic superconductors, which includes the systems with the β, β', κ, and λ types of stacking cation molecules in the conducting plane, the metallic spin correlations, the spin fluctuations of which are maximum near the wave vector (π, π), mediate superconductivity. In fact, two adjacent cation molecules form a dimer are considered as a site. Furthermore, electron hopping between a site and its nearest-neighbor sites in the transverse and longitudinal directions was described by the transfer integrals, t, and they create a type of square lattice. The second nearest neighbor sites were then determined by the diagonal transfer integral (from upper-right to lower-left corner of the square), t'. Then, the so-called anisotropic triangular lattice was characterized by the ratio, t'/t. Accordingly, the system reached frustration at $t'/t = 1$. The variation from a small t'/t (feature of the square lattice) passing through the frustration regime to a large t'/t (feature of quasi-one-dimensionality) changes the superconducting symmetry in the following manner. As the value of t'/t is varied, the superconducting symmetry change from "$d_{x^2-y^2}$" for $t'/t < 0.93$, to "$(s+d_{xy})+i(d_{x^2-y^2})$" for $t' \sim t$, to "$s+d_{xy}$" for $t'/t \gtrsim 1.3$. It should be noticed that the "$(s + d_{xy}) + i(d_{x^2-y^2})$" state breaks time-reversal symmetry according to the momentum-dependence plot of the argument of the complex number of the superconducting gap for several cases of t'/t.

We focused on the study of the superconducting state in quasi-two-dimensional organic superconductor λ-$(BETS)_2GaCl_4$ with BETS = $(CH_2)_2S_2Se_2C_6S_2S_2(CH_2)_2$ through muon spin relaxation (μSR). By substituting Se with S, which yields a negative pressure, the superconducting state eventually changed to an antiferromagnetic state.[4,5)] Our transverse-field μSR study revealed that λ-$(BETS)_2GaCl_4$ has the characteristics both d-wave and s-wave pairing symmetry.[6)] From our calculation on the basis of the density functional theory of the transfer integral within BETS dimers, λ-$(BETS)_2GaCl_4$ has two kinds of square lattices alternating in the a-direction. One has $t'/t \sim 0.8$, and the other has $t'/t \sim 0.2$.[7)] Accordingly, the lattice is not simple, reflecting its superconducting pairing symmetry. In order to investigate the possibility of time-reversal symmetry breaking in λ-$(BETS)_2GaCl_4$, we performed zero-field μSR in the superconducting state, which can sensitively detect the spontaneous appearance of an internal field of the order of about 10^{-4} T.

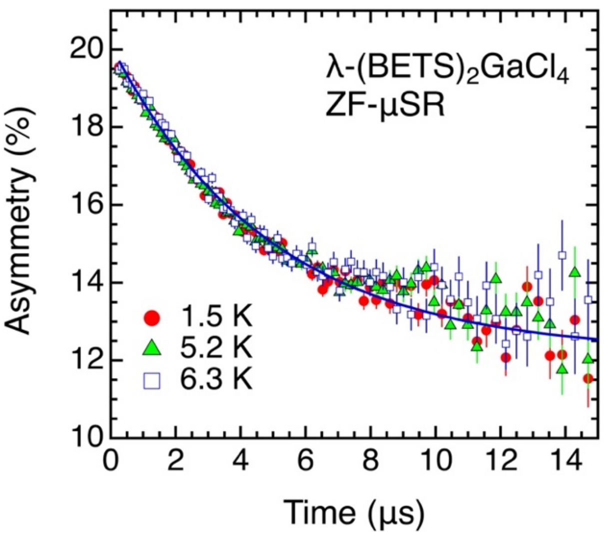

Fig. 1. Zero-field μSR time spectra measured at several temperatures, below and above T_C of $\sim$5.5 K. The solid line is a single exponential fitting model.

We prepared $\sim$130 mg single crystals of λ-$(BETS)_2GaCl_4$ and oriented them all in the same direction. The polarized muon beam direction was nearly perpendicular to the conducting plane. Zero-field μSR was conducted with high statistics up to 80 MEvents. Figure 1 shows the time spectra at several temperatures below and above the critical temperature, T_C, of $\sim$5.5 K. The time spectra up to 10 μs can fairly be analyzed using a single exponential function. No appreciable change of the relaxation rate was detected from the measurement at T = 6.3 K down to 1.5 K. We will perform similar measurements in different orientations of single crystals with respect to the muon beam direction in order to obtain complete and precise results.

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