Zero-field μSR on the out-of-plane superconductivity of λ-(BETS)$_2$GaCl$_4$

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λ-(BETS)$_2$GaCl$_4$ is a quasi-two-dimensional organic metal and it is reported as a high-anisotropic type-II superconductor. It exhibits rich physical properties once we substitute Ga with Fe, Cl with Br, or once we reduce the Se content in the BETS molecule (BETS = (CH$_2$)$_2$S$_2$Se$_2$C$_6$S$_2$S$_2$(CH$_2$)$_2$).[1)] In the P-T phase diagram, the neighboring phase adjacent to the superconducting (SC) state is the Mott insulating state, which is antiferromagnetically ordered, and separated by a suspicious paramagnetic insulating state with no magnetic ordering down to 0.3 K.[2)] A theoretical study based on anisotropic spin fluctuations-mediated superconductivity suggested an anisotropic gap structure with two nodal lines,[3)] whereas ^{13}C-NMR study evidenced two types of magnetic fluctuations of the π-electron down to 2 K.[4)] There is an urgent requirement for the detailed experimental determination of the SC gap structure. We performed a series of transverse-field (TF) μSR measurement to determine the SC gap structure. First, we found that the SC gap structure of λ-(BETS)$_2$GaCl$_4$ was unusual as it exhibited both d-wave and s-wave symmetry characteristics.[4,5)] Furthermore, considering the study by Powell and McKenzie for entire families of organic superconductors, we plan to confirm whether time reversal symmetry breaking is related to this unusual SC gap structure. Zero-field (ZF) μSR is a powerful tool to detect such breaking symmetry in the SC state.

We recently performed a series of zero-field μSR using aligned single crystals. We prepared ~130 mg single crystals and oriented them in the same direction. Measurements were performed using the ARGUS spectrometer at the RIKEN-RAL pulse muon facility with a HELIOX cryostat and fly-path setup. The single pulse mode was used to collect 80M events. In a previous report[6)] we showed the result when the polarized muon beam direction was nearly perpendicular to the conducting plane. We prepared another sample setup (~100 mg) to measure with the polarized muon beam direction parallel to the conducting plane. In this report, we compare both measurements to gain information regarding out-of-plane superconductivity.

Figure 1 shows normalized time spectra at several temperatures below and above the critical temperature (T_c ~ 5.5 K). Open circle curves indicate normalized time spectra with the muon beam direction perpendicular to the conducting plane.[6)] Blue solid lines indicate fitting lines using a stretched exponential function $A(t) = A_0 \exp(-(\lambda t)^{\beta})$. At 1.5 K, λ and β values were 0.2281(35) μs^{-1} and 1.123(16), respectively. No temperature dependence was observed between the normal and SC states. Closed circle curves indicate normalized time spectra with the muon beam direction parallel to conducting plane, which reflects the intrinsic behavior of the out-of-plane properties. Although there was no temperature dependence between the normal and SC states, we found that the shape of the spectra was distinct with the open circle spectra showing in-plane properties in λ-(BETS)$_2$GaCl$_4$. All spectra were best fit by a stretched exponential function. At 1 K, λ and β values were 1.881(21) μs^{-1} and 1.34(2). In-plane properties have a stronger relaxation while the shape is toward a single exponential function. λ-(BETS)$_2$GaCl$_4$ has high-anisotropy SC properties such as the large ratio of in-plane/out-of-plane upper critical field and transfer integral, $H_{c2||}/H_{c2\perp} > 4$[7)] and $t_{||}/t_{\perp} \sim 13$,[3)] respectively. Then, the stronger in-plane λ can be related to the antiferromagnetic fluctuations of the π-electron within the conducting plane, which mediates the superconductivity.

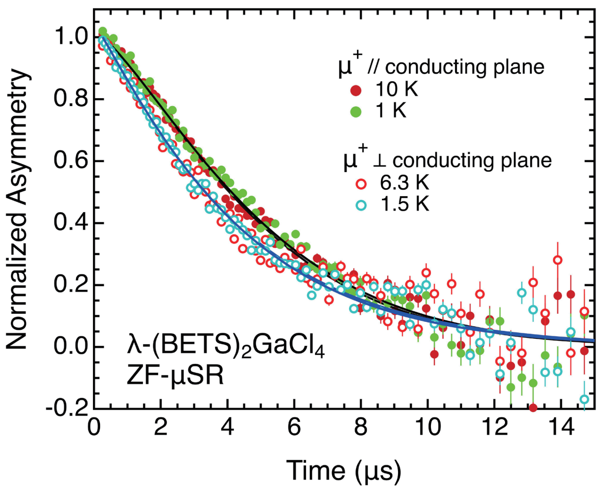

Fig. 1. Normalized ZF-μSR time spectra with the setup of μ^+ injected parallel and perpendicular to the conducting plane. The solid lines represent fitting lines obtained using the stretched exponential function.

These results confirm that the exotic SC state behavior proposed in this material based on TF-μSR could be beyond triplet superconductivity or time-reversal symmetry breaking properties. The next step is to relate this ZF-μSR result with the TF-μSR results using single crystals.

References
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