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µSR Study of Organic Superconductor λ-$(BETS)_2GaCl_4$

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Abstract. Muon-spin-relaxation (μSR) measurements in the transverse-field (TF) of 30 G were carried out from 7 K down to 1.8 K on the non-magnetic anion-based organic superconductor λ-$(BETS)_2GaCl_4$. The TF-µSR time spectrum showed a significant increase with the Gaussian-type damping behavior below the superconducting transition temperature T_C = 5 K confirming the bulk SC state with the full volume fraction. The zero-field (ZF) µSR time spectrum did not show any change against the temperature down to 1.7 K, suggesting that the time reversal symmetry of the Cooper pair might not be broken.

1. Introduction

The quasi two-dimensional organic superconductor, λ-$(BETS)_2GaCl_4$ (where BETS stands for $C_{10}S_4Se_4H_8$, bisethylenedithiotetraselenafulvalene) is metallic and undergoes the superconducting (SC) state below T_C = 5 K. Fig. 1 shows the crystal structure and a photograph of λ-$(BETS)_2GaCl_4$ (BGC). This system attracts on-going interest as this system contains non-magnetic anions. If we change the closed shell of the Ga ion to be the Fe one ion, it shows predictably similar SC properties in the field of 33 T even though the crystal sturucture does not change [1]. The first report of the specific heat measurement on BGC suggested a conventional full-gap superconductor [2]. But more recent high-resolution thermodynamic measurement by using a utilized ruthenium oxide temperature sensor, down to 0.6 K, suggesting the *d*-wave SC state [3]. Other experiments such as the flux flow resistance [4] and the scan tunneling microscopy [5] supported conventional and unconventional possibilities. One of the good methods of investigating the SC state of organic superconductors is muon spin relaxation (μSR). TF-μSR can be used to determine the magnetic penetration depth λ which related to superfluid density. The temperature dependence of $\lambda(T)$ provides an important test of the gap symmetry. This technique has been succesfully applied to studies of the previous generation of organic superconductors $(TMTSF)_2ClO_4$ and κ-$(BEDT\text{-}TTF)_2Cu(NCS)_2$ [6]. Additional advantage of using μSR is that the measurement in the zero field. An injucted muon is a sensitive probe of local internal magnetic fields of ≈ 0.1 G which corresponds to that caused by nuclear dipole moments.

Thus, it is an ideal probe to search spontaneous internal fields yielding as a result of the breaking of the time-reversal symmetry of the Cooper pair in the unconventional SC [7]. In this paper, we report a preminarily μSR study on BGC.

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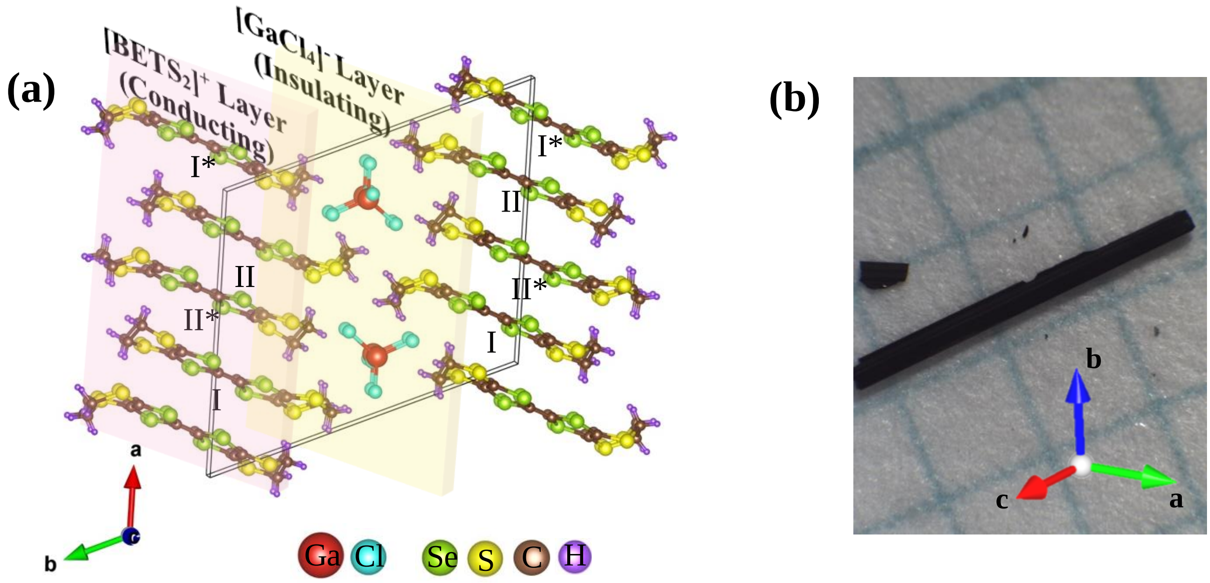

Figure 1: (a) Crystal structure of λ-$(BETS)_2GaCl_4$. There are four BETS molecules (BETS-I, I*, II and II*) in a unit cell which are containing of two types of those, Those four BETS molecules are crystallographic ally equivalent and are in the inversion symmetry with each other and in the same direction along the fourfold quasi-stacking structure. This alignment makes the crystal morphology needle-like prism. (b) Photograph of black λ-$(BETS)_2GaCl_4$ crystal. The average crystal dimensions are 3 x 0.26 x 0.13 mm^3.

2. Experimental Details

Single crystal samples of BGC were syntheiszed by the electrochemical oxidation method [8]. The zero-resisivity below 5.2 K and the diamangetization with the full volume fraction were confirmed from the resitivity and magnetic susceptibility measurements. Single crystals are crushed for μSR studies making the sample to be in the coarsed state. μSR measurements were carried out at the RIKEN-RAL Muon Facility in the UK.

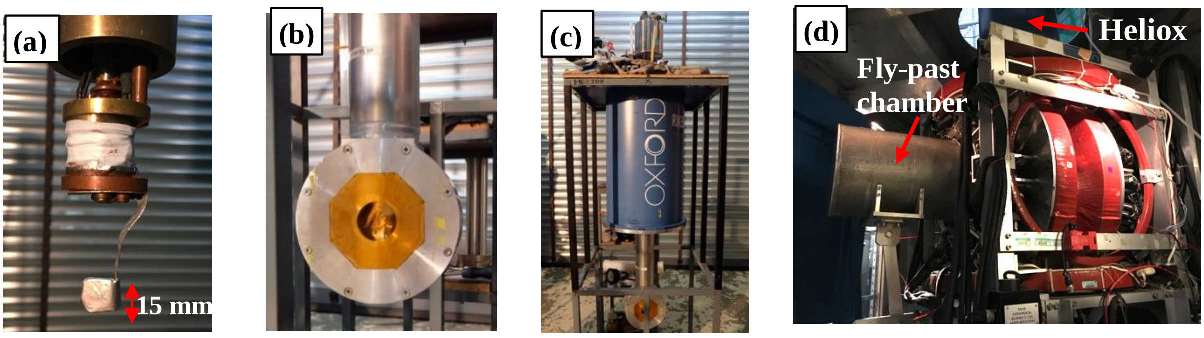

Figure 2. (a) Sample mounting. Coarsed crystals with the 120 mg mass are packed by the using 25 µm thickness and high purity (99.999 %) silver foil. The sample holder is designed for the fly-past setup for the ^{3}He cryostat. (b) The fly-past set up for the ^{3}He cryostat named HELIOX. The sample is located at the center of circular part of the tail. (c) Whole view of HELIOX with the fly-past setup. (d) The vaccume chammber extension for the fly-past setup installed on the μSR spectrometer in Port-2 of RIKEN-RAL Muon Facility, named ARGUS.

A ^{3}He cryostat was used for TF-μSR to cool the sample down to 1.8 K. Figure 2 show pictures of the experimental setup. We applied TF in perpendicular to the initial muon-spin polarization in the normal state at 7 K. And then, we did the field-cooling down to 1.8 K. A ^{4}He flow-type cryostat was used for ZF-μSR to cool the sample down to 1.7 K. The zero-field calibration was done to maintain the zero-field condition to be less than 10 mG.

3. Results and Discussions

Figure 3 shows the TF-μSR time spectrum at 7 K (in the normal state) and at 1.8 K (in the SC state). The damping behavior of the muon-spin precession due to inhomogenious internal fields at the muon site was observed. The damping behavior at 7 K is due to the distribution of local fields at the muon which is broadenend mainly by randomly oriented nuclear moments surronding the muon stopping site [9].

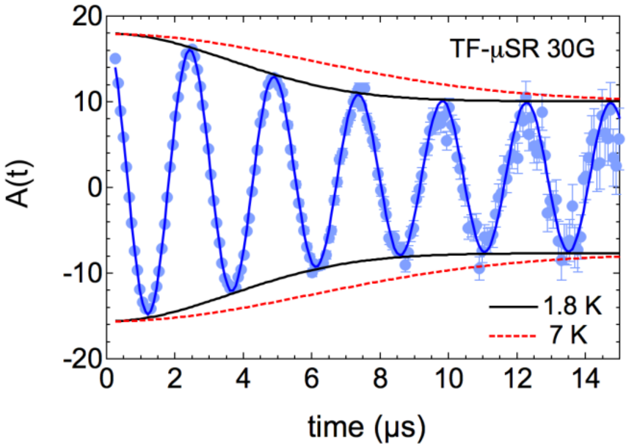

Figure 3. TF-μSR time spectrum of λ-(BETS)$_2$GaCl$_4$ under TF= 30 G at 1.8 K (close blue circle) and the best-fit result of the data by using Eq. (2). The black solid line is the damping part of the fitting result, whose rate value is σ, at 1.8 K and the broken red line is that of the one at 7 K.

We analyse the time spectra by using the following function;

$$P^{TF}(t) = A_1{}^{(-\sigma t)^2} \cos\left(\gamma_\mu H_{ext_1} t + \phi\right) + A_2 \cos(\gamma_\mu H_{ext_2} t + \phi). \quad (1)$$

Here, σ is the Gaussian damping rate, $H_{ext_1}$is the avaraged field at the muon side in the SC state. The A_1 and A_2 are asymmetry parameters of the Gayssian-type damping and the background components, respectively. The A_2 was fixed to be that achieved at the base temperature. H_{ext_2} is TF and ϕ is the phase of the muon-spin precession. In the normal state, σ was estimated to be 0.1172 + 0.0023 μs^{-1}. The Gaussian damping rate in the SC state, σ_{SC}, can be calculated from the relation $\sigma^2 = \sigma_{SC}^2 + \sigma_{NM}^2$ where σ_{NM} is the Gaussian damping rate of the background signal, and estimated to be 0.1708 + 0.0022 μs^{-1}. This decrease in the σ_{SC} compared in the SC state is caused by the appearance of the flux state which produces the distribution of the penetrated magnetic fields in the sample. The detail analysis on temperature dependence of σ_{SC} is on-going at this moment and will be publised in a separated paper.

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Figure 4(a) shows the ZF-μSR time spectra masured above/at/below T_C. No change oscillation in the time spectra was confirmed within the experimental tollerance. Time spctrra were analyzed by the streached exponential function;

$$P^{ZF}(t) = A_1\left(\exp(-\sigma t)^{\beta}\right) + A_2. \quad (2)$$

The β was estimated to be 0.88 which was close to that of the Lorentzian function. This Lorentzian-like depolarization behavior tends to be well observed in other organic superconductors [9]. No change in the time sprctrume below T_C in ZF means that no spontaneous magnetic field appeared in the SC state and the time reversal symmetry of the Cooper pair is not broken in BGC. In the case of that the Cooper pair has the *p*-wave pairing symmetry, sponteneous internal field appears in the SC state. This sponteneous internal field is small in the order of a couple of Gauss but can be picked up by the muon. Therefore, our current results can conclude that the *p*-wave pairing symmetry in inlikely to explain the SC state of BGC. More detailed μSR investigations are now going on by us at the RIKEN-RAL Muon Faclity and the results will be published very soon. [11]

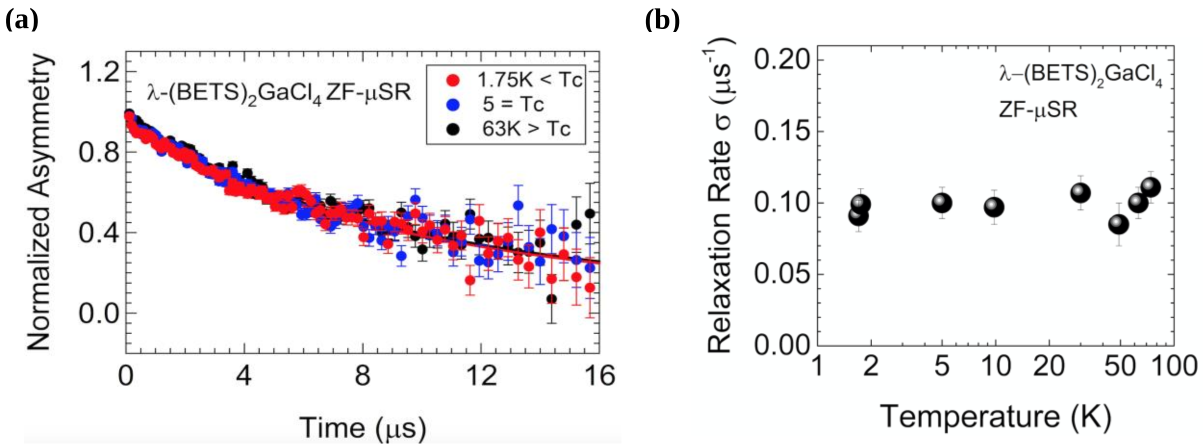

Figure 4. a) ZF- μSR time spectra at 63 K (above Tc), 5 K (at Tc) and 1.75 K (below Tc). b) Temperature independence of the relaxation rate obtained from the best fit results of time spectra by using Eq.(2).

Conclusions

ZF- and TF-μSR invesigations werep performed on the organic superconductor, λ-$(BETS)_2GaCl_4$ in order to study the superconducting state. TF-μSR under 30 G down to 1.8 K confirmed the appearance of full superconducting volume fraction with Tc = 5 K. Furthermore, from ZF- μSR, the time reversal symmetry of the Cooper pair was confirmed not to be broken reducing a possibility of the *p*-wave pairing symmetry.

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