Study of muon spin rotation of the superconducting state of organic superconductor \(\lambda-(BETS)_2\text{GaCl}_4\)

D. P. Sari, R. Asahi, K. Hiraki, Y. Ishii, T. Nakano, Y. Nozue, and I. Watanabe

The Cooper pairing symmetry of the third-generation organic superconductor \(\lambda-(BETS)_2\text{GaCl}_4\) has attracted interest owing to the strongly correlated nature of this system lying near the Mott insulating phase. A recent high-resolution thermodynamic measurement reported \(d\)-wave pairing symmetry \(^2\) whereas other experiments such as microwave conductivity measurement reported \(s\)-wave. \(^3\) We report our experimental result of zero field (ZF) and transverse field (TF) \(\mu\)SR in the fields of 30 G down to 0.3 K at the RIKEN-RAL Muon Facility in the UK. From ZF-\(\mu\)SR, a slight increase in the muon-spin relaxation rate was observed below \(T_C = 5\) K indicating a signature of the appearance of an unconventional SC state. The TF-\(\mu\)SR time spectrum at the base temperature of 0.3 K shows a damping behavior in comparison with that of the one at the normal state at 10 K owing to the appearance of the flux state, which produces a distribution of penetrated magnetic fields in the sample, as shown in Fig. 1.

![Fig. 1. TF-\(\mu\)SR time spectra of \(\lambda-(BETS)_2\text{GaCl}_4\) in TF = 30 G at 0.3 K and 10 K. The red and blue dashed line are the results of fitting the data with Eq. (1).](image)

We analyse the time spectra by using the following function

\[
\rho^\text{TF}(t) = A_1 e^{-(\sigma^2 + \phi^2) \cos(\sqrt{\sigma^2/\phi^2} H_i t) + \phi} + A_2 \cos(\sqrt{\sigma^2/\phi^2} H_i t) + \phi)
\]

Here, \(\sigma\) is the Gaussian damping rate representing the symmetric field distribution in the vortices felt by muons. The \(H_i\) is the averaged field at the muon site in the sample and \(H_2\) is the one in the Ag foil. \(A_1\) and \(A_2\) are initial asymmetry parameters of the Gaussian-type damping and the background components, respectively. \(A_2\) was fixed to be that achieved at 0.3 K, and \(\phi\) is the phase of the muon-spin precession. In the normal state, \(\sigma\) was estimated to be 0.1172 \(\pm 0.0023\) \(\mu\)s\(^{-1}\).

The Gaussian damping rate in the SC state at 0.3 K, \(\sigma_{\text{SC}}\), is estimated to be 0.1708 \(\pm 0.0022\) \(\mu\)s\(^{-1}\) by the relation \(\sigma^2 = \sigma_{\text{SC}}^2 + \sigma_{\text{NM}}^2\) where \(\sigma_{\text{NM}}\) is the Gaussian damping rate by the nuclear moment in the normal state. For a polycrystalline sample and large Ginzburg-Landau parameter \(\kappa\gg 70\), \(\sigma_{\text{SC}}\) is related to the superconducting penetration depth by the relation \(\sigma_{\text{SC}} = \sqrt{\frac{0.00371}{\Phi_0/\lambda^2}}\), where \(\Phi_0 = 2.07 \times 10^{13}\) is the flux quantum.

We could find a \(d\)-wave pairing contribution in the power-law fitting of \(\lambda\) as a deviation from that expected for a typically full-gap \(x\)-wave pairing, power-value of which is 4. Furthermore, \(\lambda(T)\) curves can be fit in the clean limit using the following expression \(^4\):

\[
\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + \frac{1}{\pi} \int_0^{\infty} \frac{\rho^2}{\sqrt{E}} dE \frac{\sin(E\Phi)}{E \sqrt{E^2 - 2^2(\Phi^2)}}
\]

where \(f = [1 + \exp(E/k_B T)]^{-1}\) is the Fermi function and the temperature dependence of the gap is approximated by \(\Delta(T) = \Delta(0) \tanh[1.82(1.018(T_C/T - 1))^{0.51}]\). The angular function \(\Delta(\phi) = \Delta_0\) in the \(s\)-wave model and \(\Delta(\phi) = \Delta_0 \cos(2\phi)\) in the \(d\)-wave model, where \(\Delta_0\) is constant. The curve can be well fit by the \(s\)-wave model. Interestingly, the data just below the \(T_c\) down to about 3 K well follow the fitting of the \(d\)-wave model and show a close agreement with the extracted critical temperature \(T_c = 5.1(1)\) K. Furthermore, we attempted to fit the data by using a simple superposition of a single-gap \(s\)-wave and single-gap \(d\)-wave, as shown in Fig. 2, in order to study if there is any hint of coexistence of \(s\) and \(d\)-wave. Further experiments on TF-\(\mu\)SR under different fields and with higher statistics may be needed to make the current experimental results convincing.

References