

Superconducting gap symmetry in organic superconductor λ -(BETS) $_2$ GaCl $_4$ studied by μ SR with DFT

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The superconducting gap symmetry determination in λ -(BETS) $_2$ GaCl $_4$ has been intriguing, since the superconducting state of this organic superconductor may be linked to that of the isostructural compound, λ -(BETS) $_2$ FeCl $_4$, showing a field-induced superconductivity in the fields > 17 T.¹⁾ We have performed transverse field μ SR in fields of 150 Oe down to 0.3 K at ISIS Muon Facility in the UK. The temperature dependence of superfluid density was best described by the $s+d$ -wave with a dominant s -wave component.²⁾ Motivated by the μ SR result, we have performed the first-principles electronic structure calculations with the generalized-gradient approximation on the basis of the density functional theory (DFT). The VASP software package adopting the plane-wave basis set with cutoff energies of 500 meV was used. The ground state charge densities were computed using $4 \times 4 \times 4$ k -point sampling and crystal structure information from Ref. 3) was used. Furthermore, we constructed maximally localized Wannier orbitals on BETS dimers to make the tight-binding energy band (dimer model) reproduce the band structure of DFT based on the experimental crystal data. The calculation was done by using the HOKUSAI RIKEN supercomputer.

Figure 1 shows the DFT band energy of λ -(BETS) $_2$ GaCl $_4$. Since the system has four BETS molecules or two BETS dimers, contributing to the electronic properties, in one unit cell, there exist two bands close to the Fermi level. Those two bands were well reproduced by the dimer model with the parameter transfer integral, in the unit of meV, as follows: $t_c = 64$, $t_{d1} = 52$, $t_{d2} = 13$, $t_{d3} = 76$, $t_{d4} = 64$, $t_e = -17$. Accordingly, the Fermi surface was consisting of an open sheet and a closed pocket as shown in the bottom right panel of Fig. 1.

The superconducting gap was then calculated by using the Random Phase Approximation (RPA) method following the extended Hubbard model,

$$H = \sum_{\langle i,j \rangle} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_i U n_{i\uparrow} n_{i\downarrow} + \sum_{\langle i,j \rangle, \sigma, \sigma'} V_{ij} n_{i\sigma} n_{j\sigma'}, \quad (1)$$

where t is the transfer integral, and U and V are intra- and inter-dimer interactions. We assumed a spin-fluctuation-mediated superconductor in the spin singlet channel. $U = 0.2$ eV was introduced in the calculation in order to investigate the low-energy interaction on the BETS dimer.⁴⁾ The result is shown in Fig. 2 showing that the red colored area is the result from RPA. More-

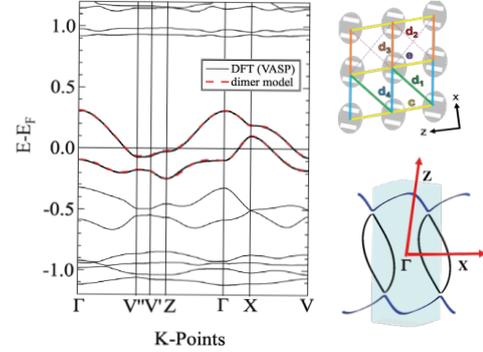


Fig. 1. DFT band structures of λ -(BETS) $_2$ GaCl $_4$. The tight-binding band structure using a dimer model with parameter inter-dimer transfer integral $t_c - t_e$ is shown as a broken line. The upper right panel shows a 2×2 molecular arrangement of the a - c conducting plane. The grey ellipse represents a BETS dimer and c - e represents inter-dimer transfer integral. The bottom right panel is the Fermi surface.

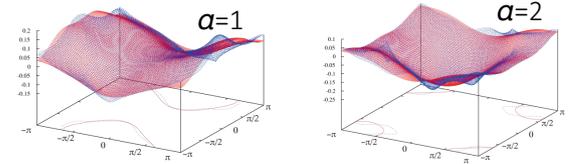


Fig. 2. Superconducting gap function from the band indexes 1 and 2, which are bands from the open sheet and closed pocket Fermi surface, respectively. The red and blue colored areas denote the RPA result and fitting result, respectively.

over, the pairing symmetry from RPA was then fitted to a linear combination of defined pairing symmetries in the k -space, with the basis functions as follows:

$$\begin{aligned} \text{isotropic } s\text{-wave: } & f_{s0}(k_x, k_y) = 1 \\ \text{extended } s\text{-wave: } & f_{s1}(k_x, k_y) = 2[\cos(k_x) + \cos(k_y)] \\ d_{x^2-y^2}\text{-wave: } & f_{d_{x^2-y^2}}(k_x, k_y) = 2[\cos(k_x) - \cos(k_y)]. \quad (2) \end{aligned}$$

The fitting result is shown in the blue-colored area of Fig. 2. We found that there exists a finite $d_{x^2-y^2}$ -wave component in conjunction with s -wave components from the contribution of two bands mentioned above. However, this mixture symmetry of $s + d_{x^2-y^2}$ indicates only one order parameter since the superconducting gap looks almost continuous along the Fermi surface, as shown in Fig. 2. Thus, from the μ SR and DFT calculation we assign a new type of superconducting gap symmetry in λ -(BETS) $_2$ GaCl $_4$, $s + d$ -wave.

References

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