PHYSICAL REVIEW B **102**, 121106(R) (2020)

Rapid Communications

Unconventional superconductivity in λ-(BETS)$_2$GaCl$_4$ probed by ^{13}C NMR

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(Received 14 May 2020; revised 3 August 2020; accepted 1 September 2020; published 16 September 2020)

We performed ^{13}C-NMR measurements on an organic superconductor λ-(BETS)$_2$GaCl$_4$ to investigate the superconducting (SC) gap symmetry, where BETS stands for bis(ethylenedithio)tetraselenafulvalene. As the temperature approaches 0 K, the Knight shift in the SC state decreases; the nuclear spin-lattice relaxation rate $1/T_1$ shows cubic temperature dependence below the SC transition temperature T_c without a coherence peak. These results can be understood in terms of d-wave superconductivity. Moreover, an increase in $1/T_1T$ at lower temperatures was observed just above T_c; this behavior can be interpreted as an effect of spin-density-wave (SDW) fluctuation. We suggest that the d-wave superconductivity of λ-(BETS)$_2$GaCl$_4$ originates from SDW fluctuation.

DOI: 10.1103/PhysRevB.102.121106

Most organic superconductors possess low-dimensional crystal structure, and the superconducting (SC) phase is situated in the vicinity of the antiferromagnetic insulating phase owing to the competition between bandwidth and Coulomb repulsion [1]. Since these features bring about exotic SC properties and are common to copper oxide superconductors, organic superconductors have also been extensively studied as strongly correlated electron systems [2,3]. Among them, the organic conductors λ-(BETS)$_2M$Cl$_4$ [BETS = bis(ethylenedithio)tetraselenafulvalene; M = Fe, Ga] exhibit fascinating SC phenomena. λ-(BETS)$_2$GaCl$_4$ exhibits SC transition at $T_c \sim 6$ K at ambient pressure [4–6]. The occurrence of Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconductivity in the vicinity of the upper critical field has been proposed [5–7]. Although λ-(BETS)$_2$FeCl$_4$ shows metal-insulator transition at 8 K [8], superconductivity is induced by external pressure [9] and magnetic field [10], and there is an FFLO phase adjacent to the field-induced SC phase [11].

To understand their exotic SC properties and their mechanism, investigation on the symmetry of the SC gap and the spin state of the Cooper pair is required. Such mechanistic discussion is already extensive for superconductors, such as cuprates, ruthenates, and heavy fermions [12]. For λ-(BETS)$_2$GaCl$_4$, there are several reports on the symmetry of the superconductivity. Recent heat capacity measured down to 0.6 K reported that λ-(BETS)$_2$GaCl$_4$ has a line-nodal SC gap [13,14]. Moreover, d-wave superconductivity was indicated via scanning tunneling microscopy and angle-dependent magnetoresistance experiments [15,16]. In contrast, μSR experiments suggested the presence of a mixed s- and d-wave SC gap [17] and microwave conductivity measurements suggested an s-wave SC gap [18]. Thus, sufficient consensus has not been reached on SC gap symmetry, and further research is needed.

Nuclear magnetic resonance (NMR) technique plays an important role in determining the SC gap symmetry, as established in studies of copper oxide and heavy-fermion superconductors [19,20]. Based on the Knight shift measurement, we can directly discuss spin susceptibility without the diamagnetic signal contribution, revealing the symmetry of the spin part of the Cooper pair wave function. The spin-lattice relaxation time T_1 measurement reveals the existence of SC gap nodes, which are related to the orbital part of the wave function. Despite such advantages of the NMR method, there has been no NMR study of the SC state for λ-type BETS superconductors. In the study of organic conductors, the ^{13}C-NMR technique is effective for investigating superconductivity because of the sharp NMR linewidth and high electron density. However, selective isotope substitution is necessary because there are many carbon sites in an organic molecule, and the synthesis of BETS molecules is more difficult than that of ET [ET = bis(ethylenedithio)tetrathiafulvalene] molecules. For these reasons, ^{13}C NMR in BETS-based organic superconductors has rarely been measured. Recently, we have succeeded in conducting ^{13}C-NMR measurements for λ-(BETS)$_2$GaCl$_4$ [21], and in this Rapid Communication, we report the ^{13}C-NMR results for λ-(BETS)$_2$GaCl$_4$ in the SC state and discuss the SC gap symmetry.

Single crystals of λ-(BETS)$_2$GaCl$_4$ were synthesized by standard electrochemical method [4]. ^{13}C-NMR measurements were performed using BETS molecules wherein only one side of the central C=C was substituted with ^{13}C nucleus in accordance with literature [21]. This technique is critical for accurately estimating the Knight shift because

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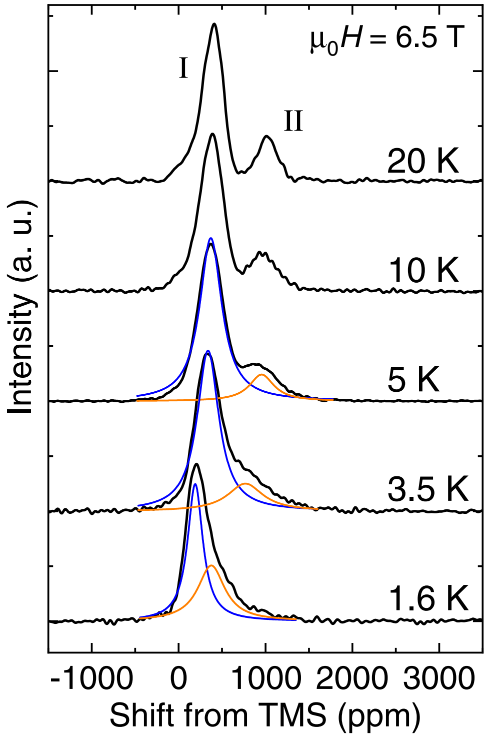

FIG. 1. NMR spectra of λ-$(BETS)_2GaCl_4$ at several temperatures in a 6.5 T field parallel to the conduction plane. Blue and yellow lines are fitting curves using Lorentzian functions.

double-side substitution causes modulation of the spectrum via dipole interaction (Pake doublet) [22], which disturbs the evaluation of the Knight shift. In a typical organic superconductor κ-$(ET)_2Cu[N(CN)_2]Br$, for which the NMR measurements in the SC state have been thoroughly performed [23–25], the magic angle configuration that can cancel the Pake doublet effect happens to coincide with the direction of the in-plane magnetic field. Hence, ^{13}C-NMR experiment via double-side ^{13}C replacement is acceptable. In the case of λ-$(BETS)_2GaCl_4$, however, there is no direction that satisfies the two conditions, and single-side substitution is important.

NMR experiments were performed on single crystals of λ-$(BETS)_2GaCl_4$ by applying magnetic fields of $\mu_0H = 2.5$ and 6.5 T parallel to the conduction plane. The field orientation was determined by measuring the resonance frequency of the NMR tank circuit at 2 T and at 2.5 K, while rotating the sample around the *c* axis using a piezoelectric rotator (within an accuracy of the relative angle ∼0.1°). Near the parallel-field condition, resonance frequency sharply decreases due to the lock-in effect [26]. NMR spectra were obtained by fast Fourier transform of the spin-echo signals following a $\pi/2$-π pulse sequence, where a typical $\pi/2$ pulse length was 2 μs. We fitted NMR spectra using Lorentzian functions, and shifts were determined with respect to the resonance frequency of the ^{13}C-NMR signal of tetramethylsilane (TMS). T_1 was measured by the conventional saturation-recovery technique.

Figure 1 shows the temperature evolution of NMR spectra obtained at $\mu_0H = 6.5$ T. At 20 K, two peaks labeled I and II were observed. In λ-$(BETS)_2GaCl_4$, two crystallographically independent BETS molecules are present, each of which has two ^{13}C sites, for a total of four ^{13}C sites. As discussed in a previous paper [21], peak I is interpreted as the superposition of three sites because the intensity of peak I is roughly three times that of peak II. As temperature decreased below 5 K, the two peaks merged and the NMR shift decreased.

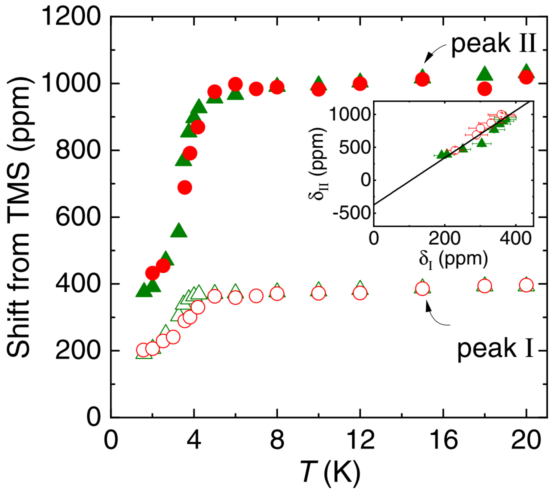

FIG. 2. Temperature dependence of NMR shift for $\mu_0H = 2.5$ T (red circles) and 6.5 T (green triangles). The inset shows δ^{I} vs δ^{II} below T_c. Solid line represents the linear fit.

To clarify the change in the NMR shift, temperature dependences of the NMR shifts of peaks I (open symbols) and II (closed symbols) are plotted in Fig. 2. Circles and triangles depict the NMR shift at $\mu_0H = 2.5$ and 6.5 T, respectively. Above 6 K, we observed nearly a temperature-independent NMR shift, indicating paramagnetic behavior. A sudden decrease was observed below $T_c = 5.1$ (2.5 T) and 3.9 K (6.5 T), consistent with the T_c estimated from the magnetic field–temperature phase diagram [5,6].

To evaluate the spin susceptibility from the NMR shift and check whether it becomes zero at low temperatures, we need to estimate the NMR shift when the spin susceptibility is zero, that is, the chemical shift. In many cases, a calculated value or estimated value from another material is used, leaving the interpretation uncertain. Hence, we determined the chemical shift experimentally as follows. NMR shift δ^i (*i*: peak I or II) can be written as follows:

$$\delta^i = K^i + \sigma = A^i\chi + \sigma, \tag{1}$$

where K^i, σ, A^i, and χ are the Knight shift, chemical shift, hyperfine coupling constant, and spin susceptibility, respectively. To discuss the contribution of the Knight shift, we need to subtract the chemical shift term. Using Eq. (1) for peaks I and II, we obtained the following equation:

$$\delta^{\mathrm{II}} = \frac{A^{\mathrm{II}}}{A^{\mathrm{I}}}\delta^{\mathrm{I}} + \left(1 - \frac{A^{\mathrm{II}}}{A^{\mathrm{I}}}\right)\sigma. \tag{2}$$

As shown in the inset of Fig. 2, δ^{II} is plotted against δ^{I} with error bars defined by a tenth of the linewidth. From the least-square fitting considering both errors of δ^{I} and δ^{II} [27], we can roughly estimate $A^{\mathrm{II}}/A^{\mathrm{I}} = 3.6(4)$ and $\sigma = 145(55)$ ppm. Both δ^{I} and δ^{II} approach the value of chemical shift toward $T = 0$ K. This result demonstrates that the spin susceptibility

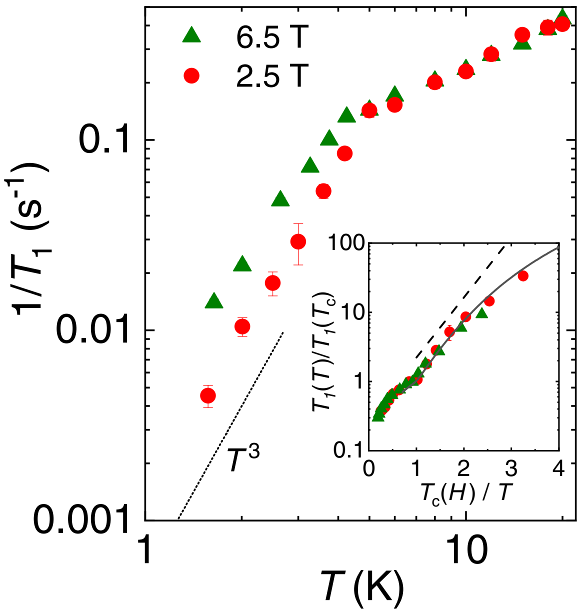

FIG. 3. Temperature dependence of $1/T_1$. Dotted line is a T^3 dependence. The inset shows an Arrhenius plot of $T_1(T)/T_1(T_c)$ vs $T_c(H)/T$. Solid curve is the calculation results assuming a two-dimensional d-wave SC gap and $2\Delta_0(0)/k_BT_c \sim 4$ using Eq. (3). Broken line is an exponential dependence indicating fully gapped superconductivity.

goes to zero, indicating the spin-singlet character of the pairing state.

The T_1 measurement provides information on the SC gap structure because it probes the shape of the density of states in the SC state. Figure 3 shows the temperature dependences of $1/T_1$ in $\mu_0H = 2.5$ and 6.5 T. In the paramagnetic state, $1/T_1$ is roughly proportional to temperature, although an increase in $1/T_1T$ with decreasing temperature can be seen in the plot of $1/T_1T$ vs temperature (Fig. 4), as discussed later. Below T_c as determined by Knight shift measurements, $1/T_1$ for both 2.5 and 6.5 T decreases in proportion to T^3 without the indication of a coherence peak, indicating that λ-(BETS)$_2$GaCl$_4$ has an anisotropic SC gap with line nodes.

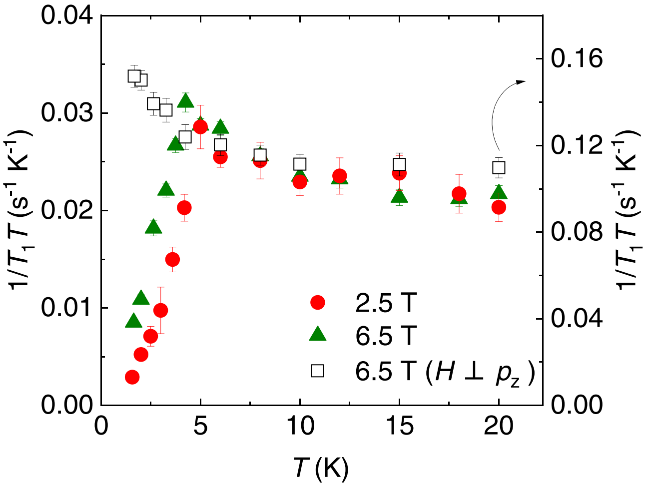

FIG. 4. Temperature dependence of $1/T_1T$ for μ_0H parallel to the conduction plane (left axis) and for μ_0H perpendicular to the p_z orbital of BETS molecule (right axis) [21].

From the Arrhenius plot of $\frac{T_1(T)}{T_1(T_c)}$ against $T_c(H)/T$ as shown in the inset of Fig. 3, we can confirm that the SC gap does not fully open among the Fermi surface. For a full-gapped superconductor, a thermally activated temperature dependence would be expected. However, $\frac{T_1(T)}{T_1(T_c)}$ deviates from the straight broken line proportional to $\exp(\Delta/k_BT)$ with $2\Delta/k_BT_c = 4$ as $T_c(H)/T$ increases, where k_B and Δ are the Boltzmann constant and the magnitude of the SC gap, respectively.

Based on these experimental results, we discuss the symmetry of the SC gap of λ-(BETS)$_2$GaCl$_4$. The Cooper pair wave function is represented by the product of the spin and orbital parts. This wave function must be antisymmetric with respect to particle exchange to satisfy the Fermi statistics. The Knight shift results for λ-(BETS)$_2$GaCl$_4$ revealed the spin-singlet state in the SC state, that is, an antisymmetric spin part of the wave function. In this case, the orbital part is symmetric, where the angular momentum of Cooper pair is even; $L = 0, 2, \ldots$ (s wave, d wave,...). Temperature dependence of $1/T_1$ exhibits T^3 dependence without a coherence peak, indicating that λ-(BETS)$_2$GaCl$_4$ is not an s-wave superconductor. Therefore, we suggest that d-wave superconductivity is the most favorable SC gap structure to explain the NMR results.

To obtain additional quantitative information, we examine the temperature dependence of $1/T_1$ in the SC state using a simple model. For a SC gap possessing line nodes, $\frac{T_1(T_c)}{T_1(T)}$ is written as [28]

$$\frac{T_1(T_c)}{T_1(T)} = \frac{2}{k_BT}\int_0^\infty \left[\frac{N_s(E,T)}{N_0}\right]^2 f(E,T)[1-f(E,T)]dE, \tag{3}$$

where $f(E,T)$ is the Fermi distribution function and $N_s(E,T) = \frac{N_0E}{\sqrt{E^2-\Delta(T)^2}}$ is the density of states in the SC state; N_0 is the density of states in the normal state and $\Delta(T)$ is the SC gap that has a temperature dependence expected in the BCS theory. As shown in the inset of Fig. 3, experimental results can be reproduced by assuming d-wave SC gap symmetry $\Delta(T) = \Delta_0(T)\cos(2\phi)$ and $2\Delta_0(0)/k_BT_c \sim 4$, where ϕ is the azimuthal angle in k space. From these results, we can also confirm d-wave superconductivity. The value of $2\Delta_0(0)/k_BT_c$ is consistent with that estimated from thermodynamic investigation [13] and suggests that λ-(BETS)$_2$GaCl$_4$ is in the weak-coupling limit.

The d-wave superconductivity result obtained here is consistent with the theoretical research by Aizawa *et al.* They considered the spin fluctuations as a pairing mechanism and proposed that Fermi surface nesting leads to a d-wave SC gap [29]. To investigate the effects on spin fluctuations, $1/T_1$ probes not only the SC gap structure but also the low-energy spin excitation in the paramagnetic state. Figure 4 shows the temperature dependences of $1/T_1T$ of the present results for μ_0H parallel to the conduction plane (left axis) and of previous results [21] for $\mu_0H = 6.5$ T perpendicular to the p_z orbital of the BETS molecule (right axis), where superconductivity is suppressed by a magnetic field. $1/T_1T$ can be written as $1/T_1T \propto \Sigma_{\mathbf{q}}|A_\perp(\mathbf{q})|^2\chi''(\mathbf{q})$ [30], where $A_\perp(\mathbf{q})$ and $\chi''(\mathbf{q})$ are, respectively, the hyperfine coupling constant under the magnetic field perpendicular to the quantization axis and the imaginary part of the dynamic susceptibility at the wave vector $\mathbf{q}$. The difference in absolute value between the left and right axes originates from the difference in $A_\perp(\mathbf{q})$ because it depends on the magnetic field direction. As temperature decreases below 10 K, $1/T_1T$ for μ_0H perpendicular to the p_z orbital increases. This enhancement with decreasing temperature was also observed just above T_c when μ_0H parallel to the conduction plane. Meanwhile, the Knight shift, which reflects the $\mathbf{q} = 0$ component of spin susceptibility, slightly decreased as the temperature decreased below 20 K [21]. Both results show that spin fluctuation develops with finite $\mathbf{q}$ above T_c. This behavior is characteristic in unconventional superconductors, such as high-T_c cuprate [31], heavy-fermion [32,33], and iron-based superconductors [34]. Hence, the spin fluctuation observed in λ-(BETS)$_2$GaCl$_4$ plays an important role in driving its superconductivity.

The Fermi surface nesting indicated by Aizawa *et al.* [29] is reminiscent of the possibility of spin-density-wave (SDW) fluctuation as the origin of spin fluctuation in λ-(BETS)$_2$GaCl$_4$. Indeed, the Fermi surface comprises one-dimensional flat portions and cylindrical portions; thus a nesting effect is expected [35,36]. As experimental evidence demonstrating such properties, we recently found divergent $1/T_1T$ behavior with the metal-insulator transition at 13 K, suggesting SDW ordering, in λ-(BETS)$_2$GaBr$_x$Cl$_{4-x}$ ($x = 0.75$), which is just close to the SC phase as a function of x [37]. The $1/T_1T$ enhancement observed at low temperatures observed in λ-(BETS)$_2$GaCl$_4$ can be interpreted as an effect of adjacent SDW ordering.

The relationship between SDW ordering in the vicinity of the SC phase and SDW fluctuation just above the SC transition has been investigated for (TMTSF)$_2$PF$_6$, where TMTSF denotes tetramethyltetraselenafulvalene. This compound shows divergent $1/T_1T$ behavior at 12 K due to SDW ordering at ambient pressure [38]. The application of pressure suppresses the SDW transition temperature and divergent behavior of $1/T_1T$, and at approximately 1 GPa, superconductivity appears with the remaining $1/T_1T$ enhancement at low temperatures [39,40]. The pressure dependence of the Curie constant estimated from $1/T_1T$ correlates with that of T_c, suggesting that SDW fluctuation is related to the SC mechanism [40]. In λ-(BETS)$_2$GaBr$_x$Cl$_{4-x}$, the decrease in x from 0.75 to 0 can be regarded as positive chemical pressure [41]; thus, the situation is qualitatively the same as for (TMTSF)$_2$PF$_6$. Indeed, SDW fluctuation has been discussed as a mechanism of superconductivity theoretically [42,43]. Given the above evidence, we suggest that the superconductivity of λ-(BETS)$_2$GaCl$_4$ is mediated by SDW fluctuation.

In summary, ^{13}C-NMR measurement was performed to investigate the symmetry of the SC order parameter of λ-(BETS)$_2$GaCl$_4$. In the SC state, the Knight shift decreases toward $T = 0$ K, revealing that λ-(BETS)$_2$GaCl$_4$ is in the spin-singlet state. $1/T_1$ shows T^3 dependence below T_c without a coherence peak, suggesting the existence of line nodes on the SC gap. These results suggest that the SC gap symmetry is most likely to be *d*-wave symmetry. Indeed, the temperature dependence of $1/T_1$ below T_c can be well explained by a two-dimensional *d*-wave SC gap. Furthermore, $1/T_1T$ enhancement observed just above T_c indicates the occurrence of spin fluctuation. We suggest that the pairing mechanism of λ-(BETS)$_2$GaCl$_4$ is intimately related to SDW fluctuation.

We would like to thank S. Hoshino for useful comments. This work was partially supported by the Japan Society for the Promotion of Science KAKENHI Grants No. 16K05427, No. 18H05843, and No. 19K21033.

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