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Evidence of antiferromagnetic fluctuation in the unconventional superconductor λ-$(BETS)_2GaCl_4$ by ^{13}C NMR

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We performed ^{13}C NMR measurements on the unconventional organic superconductor λ-$(BETS)_2GaCl_4$ to clarify its electronic properties in the paramagnetic state. We found that the spin-lattice relaxation rate divided by temperature $1/T_1T$ shows Curie-like enhancement above 55 K that arises from an antiferromagnetic fluctuation. In addition, we found additional enhancement of $1/T_1T$ below 10 K, where the Knight shift decreases and the linewidth of NMR spectra is broadened. The result could be understood by the magnetic fluctuation induced by the nesting of the Fermi surface. We discovered that λ-$(BETS)_2GaCl_4$ salt inherently exhibits the two different types of magnetic fluctuations.

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In unconventional superconductors, an insulating phase accompanied by magnetic or charge order has been frequently observed near the superconducting (SC) phase. The insulating phase is thought to play a crucial role in the emergence of superconductivity in high-T_c cuprates and organic superconductors [1]. To understand the properties of unconventional superconductivity, electronic properties in the adjacent insulating phase and the normal state just above the SC phase transition temperature T_c should be investigated. An organic superconductor, λ-$(BETS)_2GaCl_4$ [BETS: bis(ethylenedithio)tetraselenafulvalene], is one such unconventional superconductor. A thermodynamic study of its low-temperature properties reported that this λ-$(BETS)_2GaCl_4$ salt has d-wave SC gap symmetry [2]. Furthermore, near the upper critical field, hints of the presence of a Fulde-Ferrell-Larkin-Ovchinnikov phase have been found in several experiments [3–5].

Despite extensive studies for the superconductivity, the pressure-temperature (P-T) phase diagram has not been established in the λ modification in contrast to the well-known κ modification [6]. To examine the P-T phase diagram of the λ modification, a preliminary study was conducted wherein the bandwidth was controlled by changing the donor in a series of λ-D_2GaCl_4 (D = ET, STF, BETS) salts [7]; here ET and STF denote bis(ethylenedithio)tetrathiafulvalene and unsymmetrical-bis(ethylenedithio) diselenadithiafulvalene, respectively. The λ-$(ET)_2GaCl_4$ and λ-$(STF)_2GaCl_4$ salts are insulators whereas the λ-$(BETS)_2GaCl_4$ salt is metallic at ambient pressure. Resistivity measurements taken of the λ-D_2GaCl_4 salts under pressure revealed that the substitution effect of ET, STF, and BETS molecules can be understood in terms of chemical pressure. The electron-spin-resonance measurement for λ-$(ET)_2GaCl_4$ salt suggests an antiferromagnetic (AF) phase transition at T_N = 13 K [7]. Recently, a SC phase transition was observed in the λ-$(STF)_2GaCl_4$ salt under pressure [8]. From these results, we can obtain a P-T phase diagram [Fig. 1(a)] that plots the T_N, T_c, and semiconductor-metal crossover temperature T_{MI} cited from Refs. [7–9]. The positions of each salt in terms of pressure were estimated by comparing trends in the resistivity and the pressure dependence of T_c. From the P-T phase diagram, it is interesting to examine the effect of the AF phase on the electronic state in the high-pressure region. On the other hand, physical properties in a series of λ-$(BETS)_2GaBr_xCl_{4-x}$ ($0 \leqslant x \leqslant 2.0$) have been investigated [9]. Increase in the bromine content x causes the negative chemical pressure effect and insulating behavior is induced. In this insulating phase, magnetic properties were nonmagnetic, which suggested that the SC phase of λ-$(BETS)_2GaCl_4$ salt was adjacent to the nonmagnetic insulating phase. Therefore, we need to uncover whether the magnetic fluctuation remains in the superconducting λ-$(BETS)_2GaCl_4$ salt.

NMR spectroscopy probes the magnetic properties of materials as their local spin susceptibility and magnetic fluctuations can be observed through the Knight shift and the nuclear spin-lattice relaxation time T_1 measurements, respectively. ^1H and ^{77}Se NMR measurements have been performed previously, but several results appear contradictory. An increase in $1/T_1T$ was observed below 25 K in ^1H NMR [11], but the ^{77}Se NMR experiment suggested that $1/T_1T$ is constant or weak temperature dependent below 70 K [12,13]. Moreover, the linewidths of ^{77}Se NMR using randomly oriented samples and single-crystal samples show the different behaviors at low temperatures [12,13]. The contradictions arise from experimental difficulties with low sensitivity and broad linewidths in the ^{77}Se NMR signal and the additional relaxation mechanism from molecular motions due to weak coupling of the ^1H nuclei with the π conducting electrons. Therefore, measurements from the well-established ^{13}C nuclei are desirable [14]. In this paper, we performed these ^{13}C NMR measurements by synthesizing ^{13}C-enriched BETS molecules in which one side of the central C=C bond is replaced by ^{13}C. By using the ^{13}C-enriched sample, we can overcome the difficulties of previous NMR studies because the ^{13}C sites exhibit a sharp spectral response, high sensitivity, and large electron density at the nucleus. Hence, we can reveal the magnetic properties of λ-$(BETS)_2GaCl_4$ salt more clearly.

We prepared λ-$(BETS)_2GaCl_4$ salt using ^{13}C enriched BETS molecules (for details, see Supplemental Material [15]). The size of the sample used in NMR measurement is $6.0 \times 0.28 \times 0.23$ mm^3. A T_c of 5.8 K was confirmed from the increase in the resonance frequency of the NMR coil at zero magnetic field. The NMR experiment was conducted in

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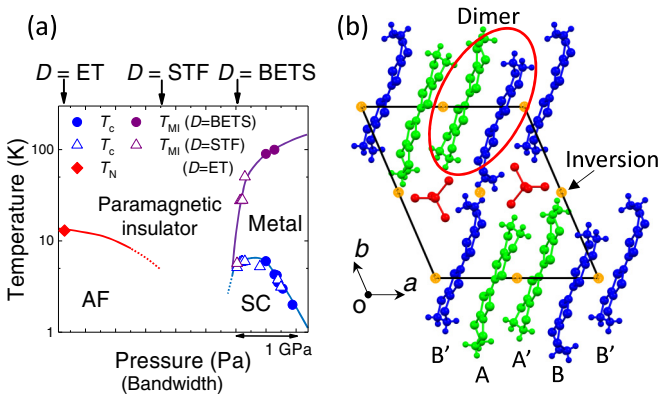


FIG. 1. (a) Temperature-pressure phase diagram of λ-D_2GaCl_4 (D = ET, STF, and BETS) salts. The data are cited from Refs. [7–9]. (b) Crystal structure of λ-$(BETS)_2GaCl_4$ salt [10]. Crystallographically nonequivalent BETS molecules A (blue) and B (green) form dimers. A′ (B′) molecules are related through inversion symmetry (yellow dots).

a magnetic field of 6.5 T. We measured the magnetic field angle dependence of NMR spectra in the a^*b^* plane (see Supplemental Material [15]). The orientation of the magnetic field θ was measured from the magnetic field parallel to the conduction plane. Temperature dependence of NMR spectra was measured at $\theta = -16^\circ$ where the peak separation of each site becomes the largest to determine the NMR shift precisely. This field orientation is perpendicular to the molecular plane. T_1 was measured at the same condition, but we could obtain the reliable data only at low temperatures because of long T_1. Therefore, the entire temperature dependence of T_1 was measured at $\theta = 56^\circ$, which enables us to obtain the high-quality T_1 data since T_1 becomes the shortest and all peaks are superposed. In these field directions, the upper critical field at 1.5 K is substantially lower than 6.5 T, so that the SC state was completely suppressed in the present NMR experiment. The NMR spectra were acquired by fast Fourier transformation of the echo signal with a $\pi/2$-π pulse sequence. Typical $\pi/2$ pulse lengths were 2 μs. The NMR shifts with respect to the reference material of tetramethylsilane and linewidths (full widths at half maximum) were evaluated by fitting the peaks to Lorentzian fitting functions. T_1 measurements were performed by the conventional saturation-recovery method.

In the λ-$(BETS)_2GaCl_4$ salt, there are two crystallographically nonequivalent molecules A and B, which have, respectively, equivalent molecules A′ and B′ related by inversion symmetry in a unit cell [Fig. 1(b)]. One BETS molecule has two crystallographically independent ^{13}C sites, so that four peaks can be expected in the NMR spectrum. Actually, we observed mainly two peaks I and II (Fig. 2). We deduce that the ratio of intensities of peaks I and II is about 3 : 1. Therefore peak I is composed of three sites. Note that a shoulder structure in the left peak can be seen. This can be understood as a consequence of the superposition of the three sites.

Figure 3(a) shows the temperature dependence of the NMR shift. The NMR shift δ^i can be written as $\delta^i = K^i + \sigma = A^i\chi_s + \sigma$ (i identifies results related to peaks I and II), where K^i, A^i, χ_s, and σ are the Knight shift, hyperfine coupling constant, spin susceptibility, and chemical shift, respectively.

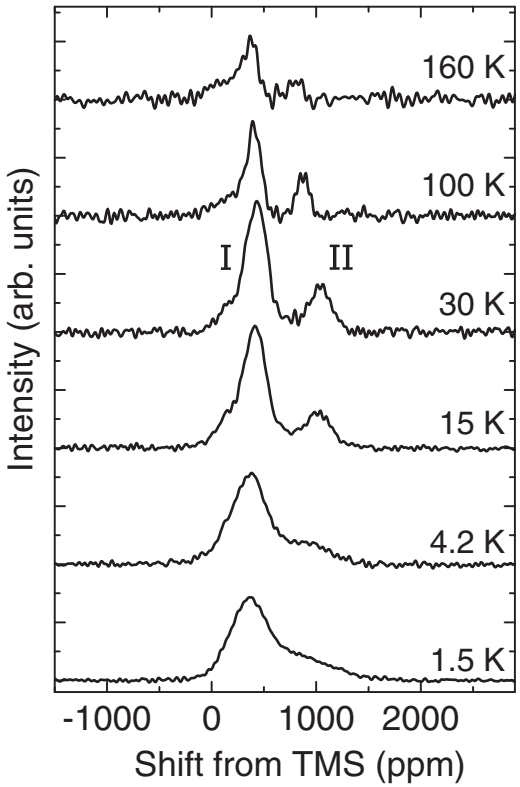


FIG. 2. NMR spectra of the λ-$(BETS)_2GaCl_4$ salt at several temperatures. There are four sites in the λ-$(BETS)_2GaCl_4$ salt, three of which overlap in peak I and the other overlaps in peak II.

To assess quantitatively the degree of spin susceptibility, we evaluated the A^i and σ by δ^i–χ plot [Fig. 3(b)]. We assume the same chemical shift for peaks I and II because the chemical shift is insensitive to the difference between A and B molecules and to the position of the central ^{13}C sites [16]. Spin susceptibility data are cited from Ref. [9]. In consequence, A^{I} for peak I is 2.61 kOe/μ_B, A^{II} for peak II is 8.55 kOe/μ_B, and σ is 149 ppm. The σ of 149 ppm is comparable to that of ET molecules [14].

The Knight shift of each spectra is also shown in Fig. 3(a) with the corresponding scale given to the right. The Knight shift weakly increases with decreasing temperature and shows peak behavior at 20 K. With a further decrease in temperature, the Knight shift decreases, where the linewidth steeply increases [Fig. 3(c)]. The linewidth at 1.5 K is approximately three times larger than that at 20 K for peak II. The linewidth which has the larger hyperfine coupling constant becomes broader at the lowest temperature. This result suggests that the line broadening comes from the inhomogeneity of the spin susceptibility. The present result of line broadening is consistent with that for ^{77}Se NMR. Moreover, we found that with decrease in temperature the line broadening is evident below 20 K, at which the Knight shift starts to decrease. To quantitatively discuss the distribution of spin susceptibility, we investigated the angle dependence of the Knight shift and linewidth (see Supplemental Material [15]). The behavior of the Knight shift and linewidth can be explained due to the development of magnetic fluctuations below 10 K. This interpretation is compatible with the

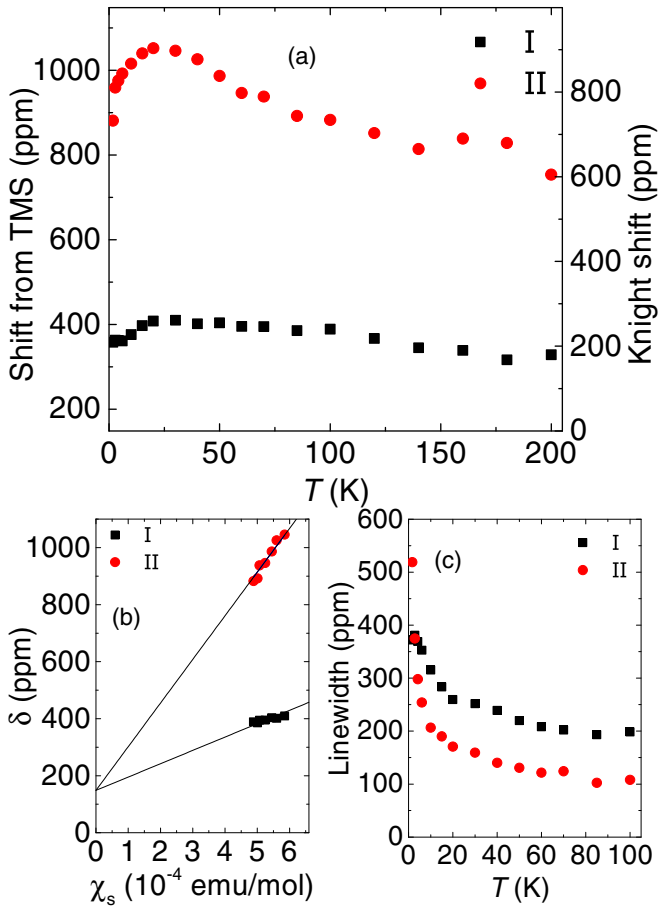


FIG. 3. (a) Temperature dependence of the NMR shift (left scale). The Knight shift, in which the chemical shift has been subtracted from the NMR shift, is shown in the right scale. (b) δ-χ_s plot between 30 and 100 K. Susceptibility data are cited from Ref. [9]. (c) Temperature dependence of linewidth below 100 K. The increasing ratio of peak I between 20 K and the lowest temperature is three times larger than that of peak II, which is ascribed to the difference in the hyperfine coupling constant.

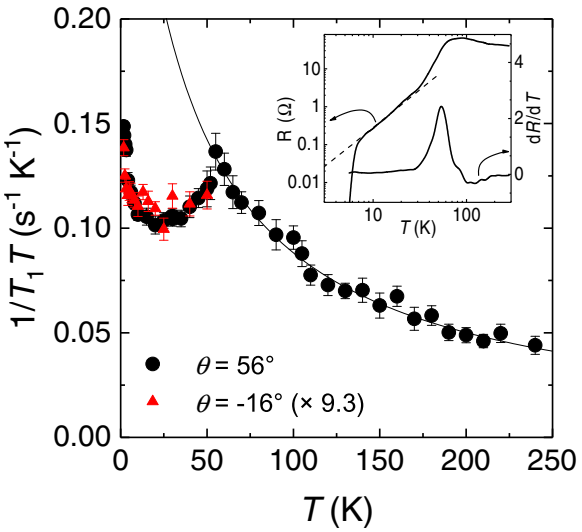


FIG. 4. Temperature dependence of $1/T_1T$ for two field directions. $\theta = -16^\circ$ is perpendicular to the molecular plane. The scale factor of 9.3 comes from the difference of the hyperfine coupling constant. The solid line plots the line $1/T_1T = C/(T+\Theta)$. The inset shows the temperature dependence of the resistivity (left scale) and its temperature derivative (right scale) [5]. The development of $1/T_1T$ suppresses below 55 K, where the resistivity indicates a semiconductor-metal crossover.

results of increase in $1/T_1T$ below 10 K as we will discuss below.

T_1 provides important information on spin fluctuations. T_1 for $\theta = 56^\circ$ is estimated from the peak which consists of four ^{13}C sites. Because each ^{13}C site has a different hyperfine coupling constant, the recovery profile deviates from the single exponential function. To correct this deviation, we used a stretched exponential function, $M(t) = M_0\{1 - \exp[-(t/T_1)^\beta]\}$. Here, $M(t)$ and M_0 are the nuclear magnetizations at t and at thermal equilibrium, and β is the stretching exponent. The recovery profiles could be fitted with β of 0.8 for all temperatures.

Figure 4 shows the temperature dependence of $1/T_1T$. $1/T_1T$ increases with decreasing temperature down to $T^* = 55$ K. Generally, $1/T_1T$ is written in terms of the summation of dynamic susceptibility $\chi''(q)$ over the wave vector q as $1/T_1T \propto \sum_q A_\perp^2(q)\frac{\chi''(q)}{\omega}$, where $A_\perp^2(q)$ and ω are the hyperfine coupling constants perpendicular to the field direction and the NMR frequency. When the system has a two-dimensional AF spin fluctuation, $1/T_1T$ can be expressed as a Curie-Weiss expression, $1/T_1T = C/(T + \Theta)$ [17]. Here C and Θ are the Curie constant and Weiss temperature. This expression is represented as the solid line in Fig. 4 with values $C = (11.6 \pm 0.4)$ s^{-1}, $\Theta = (31.5 \pm 4.2)$ K for $\theta = 56^\circ$. The temperature dependence of $1/T_1T$ above T^* is reproduced well by the Curie-Weiss expression, which suggests the existence of an AF fluctuation in λ-(BETS)$_2$GaCl$_4$ salt. Similar behavior was observed in κ-(ET)$_2$Cu[N(CN)$_2$]Br salt. $1/T_1T$ for κ-(ET)$_2$Cu[N(CN)$_2$]Br salt shows Curie-Weiss behavior above 50 K [18]. This temperature dependence of $1/T_1T$ follows that for κ-(ET)$_2$Cu[N(CN)$_2$]Cl salt which is an antiferromagnetically ordered Mott insulator. This behavior suggests that the existence of AF fluctuation in κ-(ET)$_2$Cu[N(CN)$_2$]Br salt originates from the AF Mott insulating phase. As mentioned above, the electronic phase of λ-D_2GaCl$_4$ salts can be understood using the P-T phase diagram in Fig. 1(a), which suggests that the SC phase is located near the AF phase. One candidate for the AF phase is the dimer-Mott insulator. Indeed, the intermolecular overlap integral in the dimer for the λ-(BETS)$_2$GaCl$_4$ salt is similar to that for the κ-type salt with the interplanar distances of $\sim$3.69 Å for the λ-(BETS)$_2$GaCl$_4$ salt and $\sim$3.56 Å for the κ-(ET)$_2$Cu[N(CN)$_2$]Br salt [9,19]. The tight-binding band-structure calculation for the λ-(BETS)$_2$GaCl$_4$ salt also shows this characteristic feature of a dimer-Mott system [9]. Therefore, the AF fluctuation in λ-(BETS)$_2$GaCl$_4$ salt is also derived from the AF Mott insulating phase.

At T^*, $1/T_1T$ shows a peak, below which $1/T_1T$ decreases. T^* corresponds to the temperature where the resistivity shows the inflection, as shown in the inset of Fig. 4 [5]. This result suggests that below T^*, which characterizes the crossover

from semiconductor to metal, the development of itinerancy suppresses the AF fluctuation. At around 30 K, $1/T_1T$ becomes temperature independent and the resistivity shows T^2 behavior. These are hallmarks of a Fermi-liquid state. The same relation between conductivity and AF fluctuation has been discussed in κ-$(ET)_2Cu[N(CN)_2]Br$ salt [20,21].

Whereas the electronic properties of λ-$(BETS)_2GaCl_4$ salt above 10 K are the same as those of κ-type salt, we found that $1/T_1T$ increases again below 10 K in accordance with the Curie-Weiss behavior. We can roughly estimate C and Θ as (0.12 ± 0.03) s^{-1}, $\Theta = (0.69 \pm 0.50)$ K, indicating that the origin of increase in $1/T_1T$ at low temperatures is different from the AF fluctuation above T^*. This anomalous behavior was confirmed from the temperature dependence of $1/T_1T$ on peak I for $\theta = -16^\circ$. The scale factor of 9.3 can be explained by the hyperfine coupling constant, which becomes the largest under the magnetic field perpendicular to the p_z orbital. The increase in $1/T_1T$ was also confirmable on ^{77}Se NMR measurement [13]. These results suggested that increase in $1/T_1T$ at low temperatures is intrinsic and independent of field directions. A possible explanation of increase in $1/T_1T$ is the magnetic fluctuation enhanced by the nesting effect of the Fermi surface. In fact, λ-$(BETS)_2GaCl_4$ salt has a one-dimensional sheet in the Fermi surface [22,23]. Similar Curie-like behavior of $1/T_1T$ was observed in $(TMTSF)_2PF_6$ salt under pressure [24], at which the SC state emerges near the spin-density-wave (SDW) phase. Resistivity measurements at the same pressure regions show linear temperature dependence in the normal state, destroying the superconductivity by the weak magnetic field [25], which suggests the non-Fermi-liquid state. These results indicate that the SDW fluctuation is realized in the non-Fermi-liquid state. A similar situation can be expected in λ-$(BETS)_2GaCl_4$ salt while the conducting properties have not been studied in the normal state near T_c. The existence of SDW fluctuation can consistently explain the line broadening and decrease in Knight shift. Interestingly, in the alloy of λ-$(BETS)_2GaBr_xCl_{4-x}$ salts, the salts of $x = 0.0$ to 0.75 showed the semiconductor-metal crossover behavior and a metal-insulator transition occurred at 18 K in $x = 0.75$ salt, showing the anomaly observed in the magnetic susceptibility measurement [9]. This metal-insulator transition might be the SDW transition.

Since the AF fluctuation at high temperatures is suppressed below T^*, it seems not to be related with the SC state directly. The behaviors at around T^* are reminiscent of κ-type salts, whereas the magnetic fluctuation at low temperatures can be intimately related with the SC state, suggesting the different pairing mechanism from that of κ-type salts.

In this paper, we explained the results below 20 K by the development of the magnetic fluctuation. However, ^{77}Se NMR measurement suggests that the line broadening at low temperatures originates from the charge disproportionation in the conducting layer [13], which is still the possible mechanism. Since NMR spectroscopy is sensitive not to charge but to spin, charge sensitive experiments such as infrared or Raman spectroscopies are needed to reveal the charge disproportionation.

The λ-$(BETS)_2FeCl_4$ salt exhibits a AF phase transition at zero magnetic field at 8.3 K, which has been thought to be derived from the interaction between Fe 3d spins [26]. Recently, specific-heat measurements have indicated that the antiferromagnetism in the λ-$(BETS)_2FeCl_4$ salt is caused by the AF alignment of π spins, whereas the Fe 3d spins are in the paramagnetic spin state [27]. Although the origin of AF ordering has been extensively studied, this aspect has not been settled as yet. The development of magnetic fluctuation below 10 K in the nonmagnetic λ-$(BETS)_2GaCl_4$ salt indicates that the interaction between π spins of BETS molecules is a source of AF ordering in λ-$(BETS)_2FeCl_4$ salt.

In conclusion, we prepared the ^{13}C enriched BETS molecules and performed the ^{13}C NMR measurement in λ-$(BETS)_2GaCl_4$ salt to examine the electronic properties in the paramagnetic state. The superconducting λ-$(BETS)_2GaCl_4$ salt possesses the different types of the magnetic fluctuations. Above 55 K, $1/T_1T$ shows Curie-like behavior, indicating the existence of an AF fluctuation originating from the dimer-Mott insulator. The peak behavior of $1/T_1T$ was observed at 55 K, where the semiconductor-metal crossover was observed in the electric conductivity, and shows the Fermi-liquid-like behavior at around 30 K. In addition, we also observed the increase in $1/T_1T$ below 10 K, and decrease in Knight shift and line broadening below 20 K, which can be related with the SC state. The anomaly at low temperatures can be understood by the development of the magnetic fluctuation due to the Fermi-surface nesting.

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