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Title	Evidence of antiferromagnetic fluctuation in the unconventional superconductor lambda-(BETS)(2)GaCl4 by C-13 NMR
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Supplementary information for :Evidence of antiferromagnetic fluctuations in unconventional superconductor λ-$(BETS)_2GaCl_4$ by ^{13}C NMR

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I. SAMPLE PREPARATION

BETS molecules were prepared in accordance with the prescription given in Ref. [S1]. At the last step of the synthesis of BETS molecules, 15 % of ^{13}C-enriched ketone (**1**) and 85 % of naturally abundant ketone (**2**) were mixed (Fig. S1). Thereby we obtained 72 % of C=C molecules (**3**), which is NMR inactive, 26 % of ^{13}C=C molecules (**4**), and 2 % of ^{13}C=^{13}C molecules (**5**). This coupling ratio statistically reduces the products of ^{13}C=^{13}C molecules, which cause contamination of the NMR spectrum through the Pake doublet effect[S2]. We determined the actual ratio by mass spectroscopy as 72 % of C=C molecules, 28 % of ^{13}C=C molecules, and the trace amount of ^{13}C=^{13}C molecules. Single crystals of the λ-$(BETS)_2GaCl_4$ salt were grown by the electrochemical oxidation method[S3].

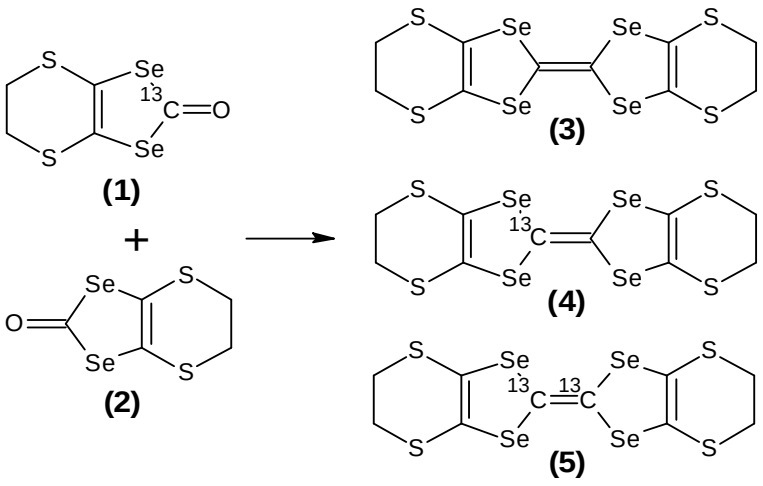


FIG. S1. Preparation of ^{13}C-enriched BETS molecules (see text).

II. FIELD ORIENTATION DEPENDENCE OF NMR SPECTRA

Field angle dependence of NMR spectra were measured at (a) 1.8 K and (b) 30 K as shown in Fig. S2. Magnetic field was rotated in the a^*b^*-plane. The orientation of the magnetic field $\theta = 0°$ corresponds to the magnetic field parallel to the conducting plane. By rotating the sample, NMR spectra change from asymmetric single peak to double peak. Figure S3 shows the field orientation dependence of NMR shift (left scale, closed symbol) and linewidth (right scale, open symbol) obtained by fitting with two Lorentzians. NMR shift and linewidth vs field orientation curves can be fitted by the sine curve. From these fittings, we can estimate that NMR shift shows the maximum at θ=−18° and shows the minimum at θ=−108°. By considering the relation between crystal structure and field orientation, the maximum (minimum) of NMR shift was observed when the magnetic field is applied perpendicular (parallel) to the molecular plane. This result is consistent with the previous ^{77}Se NMR measurements[S4]. As mentioned in the body of this article, because the chemical shift is approximately 150 ppm, main contribution of the angle dependence of the NMR shift

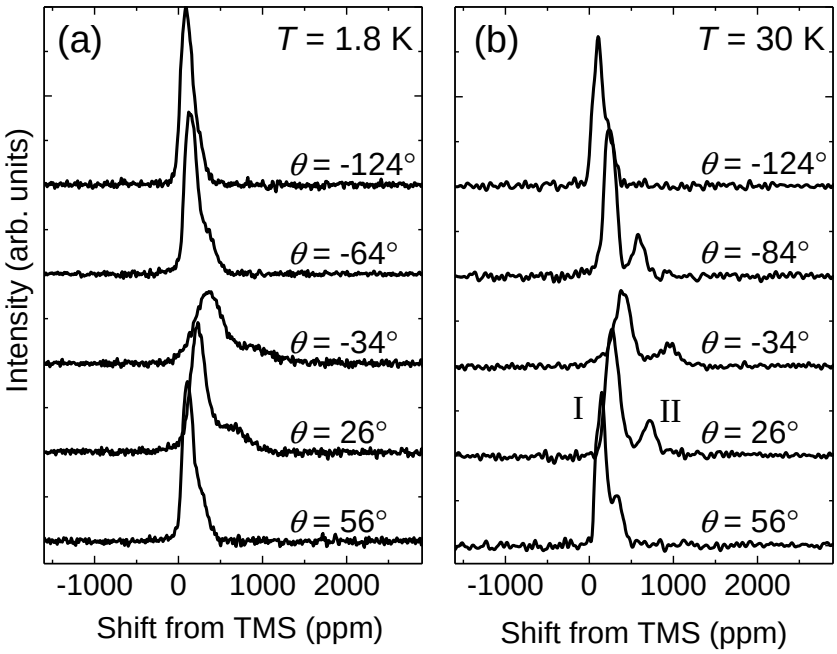


FIG. S2. Angle dependences of NMR spectra in an external magnetic field rotated in the a^*b^*-plane at (a) 1.8 K and (b) 30 K.

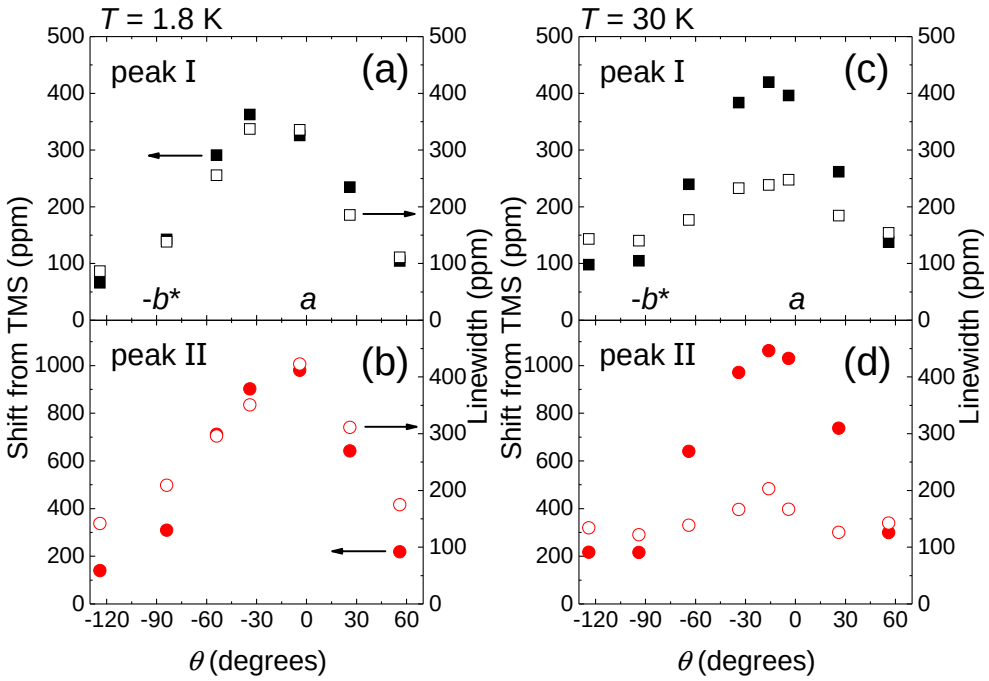


FIG. S3. Angle dependences of NMR shift (left scale) and linewidth (right scale). The origin of the horizontal axis corresponds to the magnetic field applied parallel to conduction plane.

TABLE S1. Relative distribution of the spin density $\Delta\chi/\chi$.

	1.8 K	30 K
peak I	0.91	0.33
peak II	0.29	0.061

is due to the Knight shift. Linewidth has also the same angle dependence as the Knight shift. These results suggest that the angle dependence of Knight shift $K(\theta)$ and linewidth $\omega(\theta)$ can be written as, $K(\theta) = A(\theta)\chi$ and $\omega(\theta) = A(\theta)\Delta\chi$. Here, $A(\theta)$, χ and $\Delta\chi$ are the hyperfine coupling constant, the spin susceptibility and the distribution of the spin susceptibility. To evaluate the degree of electronic inhomogeneity, we discuss the relative distribution of the spin density, $\Delta\chi/\chi = \omega(\theta)/K(\theta)$. These values of each peak are displayed in Table S1. $\Delta\chi/\chi$ for peak I is larger than that for peak II. This is consistent with the interpretation that the peak I is composed of three ^{13}C sites. Increasing ratios of peak I and II from 30 K to 1.8 K are 2.7 and 4.9, which are correlated to the hyperfine coupling constant. This result suggests that the increase in the linewidth with decreasing temperature is due to the

inhomogeneity of local spin susceptibility. We compare the results of ^{13}C NMR with ^{77}Se NMR measurements. The value $\Delta\chi/\chi = 0.29$ of peak II ($T = 1.8$ K) is almost comparable with the value $\Delta\chi/\chi = 0.3$ ($T = 1.9$ K) obtained from ^{77}Se NMR experiment[S4]. Note that the value of peal I cannot be compared quantitatively due to the superposition of three ^{13}C sites. Therefore, we suggest that the degree of inhomogeneity of local spin susceptibility is essential for λ-$(BETS)_2GaCl_4$ salt.

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