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Breakdown of the Field-Induced Superconductivity by Dynamical Spin Reversal

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The quasi-two-dimensional molecular conductor λ -(BETS)₂FeCl₄ shows a superconductivity in the high magnetic field environment. This is due to the compensation effect between the external field and the internal field created by the Fe³⁺ magnetic moment. Here, we report the simultaneous transport and electron spin resonance (ESR) measurements in the field-induced superconducting state down to 1.65 K. We find that the field-induced superconductivity is partially destroyed by ESR transitions, namely, by dynamical spin reversals of Fe³⁺. Moreover, it is found from our ESR results that the field-induced superconducting state is inhomogeneous, coexisting with the residual paramagnetic metal state.

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Keywords: Field-induced superconductor, ESR, π - d interaction, BETS

I. INTRODUCTION

For the past few decades, many attempts have been performed to include localized 3d spins in molecular conductors since interaction between the conducting π -electron and the localized 3d spin, known as the π - d interaction, yields intriguing physical properties.¹⁻⁹ The quasi-two-dimensional molecular conductors λ -(BETS)₂FeCl₄, where BETS is bis(ethylenedithio)tetraselenafulvalene, is one of the typical molecular conductors where effective π - d interaction exists in the system. As shown schematically in Fig. 1, the planar BETS molecules are arranged in the ac -plane, and consequently form two dimensional conducting layers. Meanwhile, the FeCl₄ anions lie between the BETS layers, and form insulating layers. Because of the short atomic distance between the BETS molecule and Fe³⁺ ion ($S=5/2$), considerable π - d interaction exists in the system.

Due to this specific feature, this system shows complementary physical properties both in conductivity and magnetism. At $T_N=8.3$ K, λ -(BETS)₂FeCl₄ shows metal-insulator transition associated with the antiferromagnetic long range order.^{1,2} Then, above $B_C=10$ T, the metallic state is recovered by the saturation of the canted spins.² Finally, above $B_{FISC}=17$ T, the paramagnetic metallic state undergoes to the superconducting state when the magnetic field is applied parallel to the conducting ac -plane.^{3,10}

In general, two mechanisms are responsible for the destruction of the superconducting state. One is the ‘Zeeman effect’, where the magnetic field breaks the paired electrons, and the other is the ‘orbital effect’, where the energy gain of the pair formation is lost by the penetration of the vortices into the superconductors. It is considered that, for the λ -(BETS)₂FeCl₄ system, these

destruction mechanisms are suppressed at high magnetic field for two reasons. The former effect is suppressed since the internal field created by the localized d -electrons is compensated by the external field, well known as the Jaccarino-Peter compensation effect.¹¹ Meanwhile, the latter effect can be suppressed by inducing the magnetic field parallel to the conducting plane.

These two suppression mechanisms are the key factor for the formation of the field-induced superconductivity (FISC) in the λ -(BETS)₂FeCl₄ system. Experimentally, Hiraki *et al.* have shown that huge internal field indeed exists in the system,¹² and Uji *et al.* have shown that the FISC state can be easily destroyed if the magnetic field is a few degrees off from the conducting plane.³

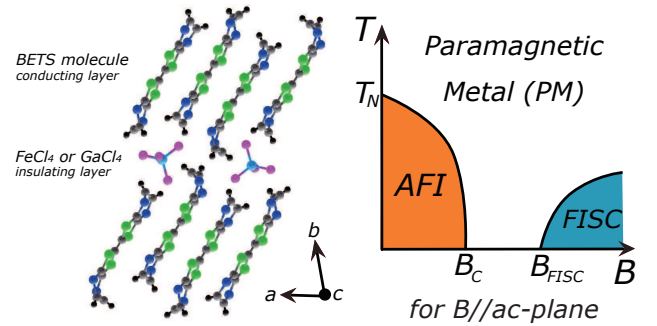


FIG. 1: (Left) Crystal structure of λ -(BETS)₂FeCl₄ salts. For mixed compound, the Fe is partially substituted to Ga. (Right) Schematic phase diagram of mixed compounds, λ -(BETS)₂Fe_{1-x}Ga_xCl₄ ($x > 0.33$). AFI and FISC denote antiferromagnetic insulator and field-induced superconductor, respectively. FISC phase appears only for $B \parallel ac$ -plane

To have a more microscopic and direct information about the role of the π - and 3d-electrons in the FISC

state, we have previously performed electron spin resonance (ESR) measurements at high magnetic field.¹³ Here we have observed a single absorption line, which suggests the existence of a strong exchange interaction between the π and d -electrons. However, we have also found that the intensity or the linewidth of the ESR signal does not change between PM state and FISC state. This was a quite surprising result since the change of skindepth, which is related to the change of the surface conductivity from PM to FISC state, normally affects the ESR intensity, and the change in the flux penetration should also affect the linewidth or the lineshape of the ESR spectra. Although we have concluded that this behavior was due to the coexistence of FISC and PM states, this result is still under discussion since a field orientation accuracy of few degrees is needed for the FISC state.³

Hence, we have built a hybrid measurement system as shown in Fig. 2, where transport and ESR can be measured simultaneously. In addition, it includes a precise rotator mechanism so that the magnetic field can be correctly applied parallel to the conducting plane, and the FISC state can be confirmed by the transport measurement. By using this system, it enables us to study the microscopic information about the electronic states of the FISC phase.

Due however, to our instrumental limitation both in magnetic field and high frequency for ESR, it is difficult to study the FISC state of a pure FeCl_4 compound, where $B_{\text{FISC}}=17$ T. Hence, we have chosen to study a mixed crystal system $\lambda\text{-(BETS)}_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ where FeCl_4 anions are partially substituted by non-magnetic GaCl_4 (see Fig. 1). By mixing these non-magnetic anions, B_{FISC} can be shifted to much lower field.

In this paper, we present the simultaneous transport and ESR measurements of $\lambda\text{-(BETS)}_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ for $x=0.34, 0.5$ and 0.6 . It is found that ESR is observed even in the FISC state suggesting an inhomogeneous superconducting state down to 1.65 K. Moreover, we have additionally found anomalies in the resistance when the ESR transition occurs. The temperature dependence and the power dependence of the millimeter-wave irradiation are presented, and the mechanism of the resistance anomaly is discussed. Consequently, it is found that the FISC state is partially destroyed due to the ESR transition, namely, the dynamical spin reversal of the Fe^{3+} ions.

II. EXPERIMENTAL

Simultaneous transport and ESR measurements have been performed at the HFLSM-IMR in Tohoku University (Sendai, Japan). The sample was prepared by the electrochemical method as described elsewhere¹⁴, and needle-shaped single crystal is obtained. The crystallographic c^* -axis corresponds to the needle axis. The typical size of one single crystal used in this study is about

$2.0 \times 0.5 \times 0.1$ (mm³).

To obtain a clear ESR signal from such tiny single crystal, we have employed the Fabry-Pérot type open cavity method as shown in Fig. 2. Millimeter wave were irradiated from 135 GHz Gunn oscillators (Millitech), which is doubled to 270 GHz by highly-efficient frequency doublers (Virginia Diodes, Inc.). The amplitude-modulated millimeter wave is guided through a lightpipe and introduced into a Fabry-Pérot resonator. The transmission is monitored by lock-in detection method using the InSb detector (QMC instruments). Note that the transmission power is optimized by changing the distance of the resonator. The sample was set on a quartz rod and positioned at the center of the resonator. The quartz rod (and sample) can be rotated precisely using a piezo rotator (Attocube Systems, ANR30). Meanwhile, four gold wires of $\phi 10$ μm are attached to the sample, and the resistance is measured simultaneously using the AC resistance bridge (Picowatt, AVS-47). The current was applied along the c^* -axis.

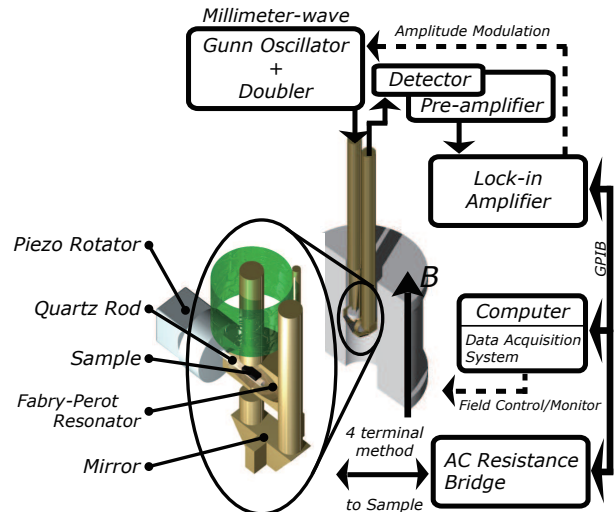


FIG. 2: Schematic diagram of the experimental setup.

III. RESULTS

A. $\lambda\text{-(BETS)}_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ ($x=0.6$)

The magnetoresistance of $x=0.6$ compound at 1.7 K for $B \perp$ and $B \parallel ac$ -plane is shown in Fig. 3(a). In association with the saturation of the magnetic moment, an insulator to metal transition is observed around 6.5 T, corresponding to B_c in Fig. 1. This transition is accompanied by a hysteresis of 0.5 T where its origin is still under discussion.^{2,15} When the magnetic field is not applied parallel to the conducting plane, a slightly positive magnetoresistance of about 0.8Ω is observed for $B > B_c$. On

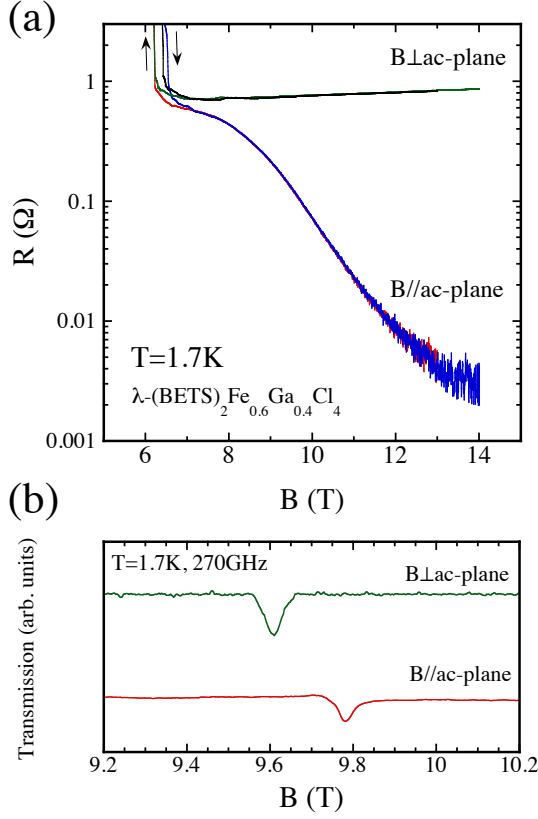


FIG. 3: (a) The magnetoresistance and (b) Typical ESR spectra of λ -(BETS) $_2$ Fe $_x$ Ga $_{1-x}$ Cl $_4$ ($x=0.6$) for $B \perp ac$ -plane and $B \parallel ac$ -plane at 1.7 K.

the other hand, for $B \parallel$ conducting ac -plane, the magnetoresistance gradually decrease as a function of magnetic field and turns into a FISC state, where the resistance is in the m Ω range.

The millimeter wave transmission of 270 GHz was recorded simultaneously with the transport measurement. Figure 3(b) shows the typical ESR spectra for $B \perp$ and $B \parallel ac$ -plane. Although a small g -shift was observed due to the rotation of the sample, single absorption lines were observed for both field orientations. The observation of a single absorption line strongly suggests that considerable π - d interaction exists in the system. It is well known, for such a case, the observed absorption predominantly originates from $3d$ spins since $S=5/2$ for Fe $^{3+}$ comparing to π -electrons which is $S=1/2$.¹⁶ Furthermore, the millimeter wave radiation cannot entirely penetrate throughout the sample. The skindepth can be written as $\delta = \sqrt{1/(\mu_0 \pi \nu \sigma)}$, where ν and σ are the frequency and conductivity, respectively. From Fig. 3(a), the conductivity at 9.8 T, where the ESR transition occurs, is about 2000 and 250 S cm $^{-1}$ for $B \parallel$ and $B \perp ac$ -plane, respectively. Then, the estimated skindepth for 270GHz is about 2 and 6 μ m for $B \parallel$ and $B \perp ac$ -plane, respectively. Therefore, the observed absorption can be attributed to the ESR coming from the Fe $^{3+}$ ions located

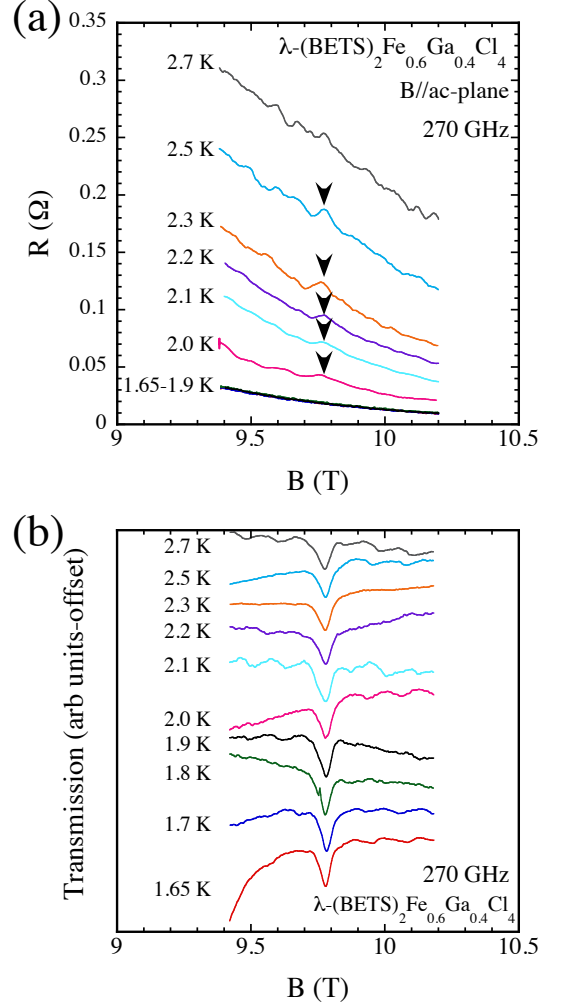


FIG. 4: The temperature dependence of (a) the magnetoresistance and (b) the ESR spectra for $x=0.6$ compound ($B \parallel ac$ -plane). The black arrows in (a) show the resistance anomalies at the resonance field

near the surface of the sample.

The ESR intensity for $B \parallel ac$ -plane (FISC phase) is smaller than $B \perp$ plane (PM phase) as shown in Fig. 3(b). Considering that the linewidth is almost identical, ca. 41.7 and 44.5 mT for $B \perp$ and $B \parallel ac$ -plane, respectively, both ESR signals should have the same origin.

There are two possible reasons for the difference of the intensities. First is the difference of the skindepth as mentioned above. The one-third skindepth reduction from $B \perp$ to $B \parallel ac$ -plane could decrease the intensity of the ESR. Second, there is still a finite resistance around 9.7 T for $B \parallel ac$ -plane in which it is supposed to be in the FISC phase. This result might suggest that the sample is only partially superconducting and PM domains are still remaining. The reduction of the PM domains from $B \perp$ to $B \parallel ac$ -plane is also a possible candidate for the decrease in the ESR intensity.

The temperature dependence of the magnetoresistance

and ESR spectra for $B \parallel ac$ -plane are shown in Fig. 4(a) and (b), respectively. Although ESR is observed down to 1.65 K, its intensity remains almost the same and does not follow a Curie law. Considering the resistance is nearly identical from 1.65 to 1.9 K, the skindepth should not vary in this temperature range, and an enhanced ESR signal should be observed at lower temperature. Hence, the observed intensity behavior is not due to the skindepth effect as mention above. It is more likely that the FISC state is inhomogeneous with the coexistence of the PM domains, and the PM domains are diminishing with decreasing temperature. This is in good agreement with the observed ESR linewidth, which does not show a drastic change with temperature. Namely, the observed ESR in the FISC state originates from the Fe^{3+} of the residual PM domains.

One striking feature in the magnetoresistance is the peak observed around 9.8 T at intermediate temperatures between 2.0 and 2.5 K as shown in Fig. 4(a) (black arrow). These resistance anomalies are observed when the ESR transition occurs, i.e. the peak is observed at the ESR's resonance field ~ 9.8 T. It is noteworthy that no peak is observed at the lowest temperature and above 2.7 K which excludes the possibility of a simple heating effect due to millimeter-wave irradiation. The mechanism of this peak is discussed in the following section. Let us also stress that this effect is observed for the first time due to the simultaneous transport and ESR measurement system.

B. $\lambda\text{-(BETS)}_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ ($x=0.5, 0.34$)

We have shown in the previous section that the ESR is observed even at the lowest temperature where the FISC state is expected, and the resistance shows a peak when ESR transition occurs in some specific temperature range. However, the resistance for $x=0.6$ compound has still finite values, and it is possible that the magnetic field is not enough for FISC state. Therefore, here in this section, we will focus on the $x=0.5$ and 0.34 compounds where FISC state is achieved in much lower field. Moreover, the power dependence of the irradiated millimeter wave is presented.

The magnetoresistance of $\lambda\text{-(BETS)}_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ ($x=0.5$) at 1.7 K for $B \parallel ac$ -plane is presented in Fig. 5. The insulator to metal transition is observed at $B_c=2.5$ T, and a smooth transition from the PM to the FISC state is observed at higher field. This result is similar with the one observed for $x=0.47$ compound¹⁷ Although the origin is still unknown, the small dip observed around 3.4 T is sometimes observed for the mixed compound.

The temperature dependence of the magnetoresistance of $\lambda\text{-(BETS)}_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ ($x=0.5$) for $B \parallel ac$ -plane in the FISC phase is shown in Fig. 6(a). One can see that huge changes in the magnetoresistance are observed as the function of temperature. The step-like structures, observed around 6 and 11 T for low and high tempera-

ture region, respectively, are probably due to the lock-in transition of vortices penetrating in the superconducting layer, which is a fingerprint of the FISC state.¹⁸

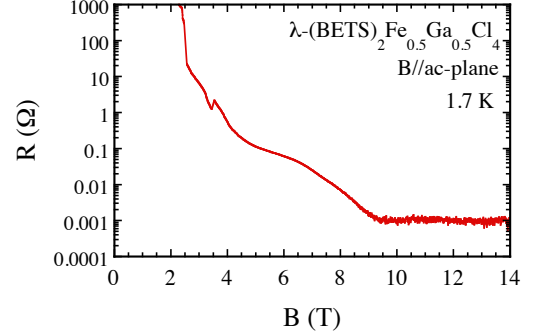


FIG. 5: The magnetoresistance of $\lambda\text{-(BETS)}_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ ($x=0.5$) at 1.7 K for $B \parallel ac$ -plane.

As shown in Fig. 6(b), the ESR signal is still observed at a temperature range where the resistance value is minimal. This is consistent with the previous $x=0.6$ results, and this suggests the observation of ESR at the FISC state do not depend on the ratio of the compound but just an intrinsic behavior of the FISC state. Note that the temperature dependence of the ESR's intensity is similar to that of the $x=0.6$ compound.

The peaks in the magnetoresistance when the ESR transition occurs are also observed for the $x=0.5$ compound (circle in Fig. 6(a)). The magnified magnetoresistance for each temperature, where the absolute value of the resistance is offset for clarity, is shown in Fig. 7(a). Initially, the peak is not observed at low temperature. However, the peak starts to appear around 2.2 K and evolves as the function of temperature, and suddenly vanishes at 4.4 K. This behavior is also similar with one observed for the $x=0.6$ compound. The observed resistance peak ΔR is evaluated from the difference between the peak at the resonance field and the background magnetoresistance, and a maximum of $\Delta R \sim 3.2$ m Ω was observed for 3.6 K.

To confirm that the resistance peak originates from the ESR transition, we have performed the power dependence of the irradiation by putting a millimeter wave attenuator at the end of the oscillator. The results for 3.6 K are shown in Fig. 7(b). The attenuation value, $10 \times \log_{10}(P_1/P_0)$, is obtained from the amplitude of the attenuated transmission P_1 versus the bare transmission P_0 . Here again, such comparison of ΔR and transmission power was only possible due to the simultaneous measurements. As shown in the inset of Fig. 7(b), ΔR decreases linearly as the function of the attenuation value, and vanishes around -10 dB. Therefore, the resistance anomaly observed at the resonance field is related to the power of the millimeter wave, namely, it is related with the ESR transition.

The results for $x=0.34$ compound, which shows FISC

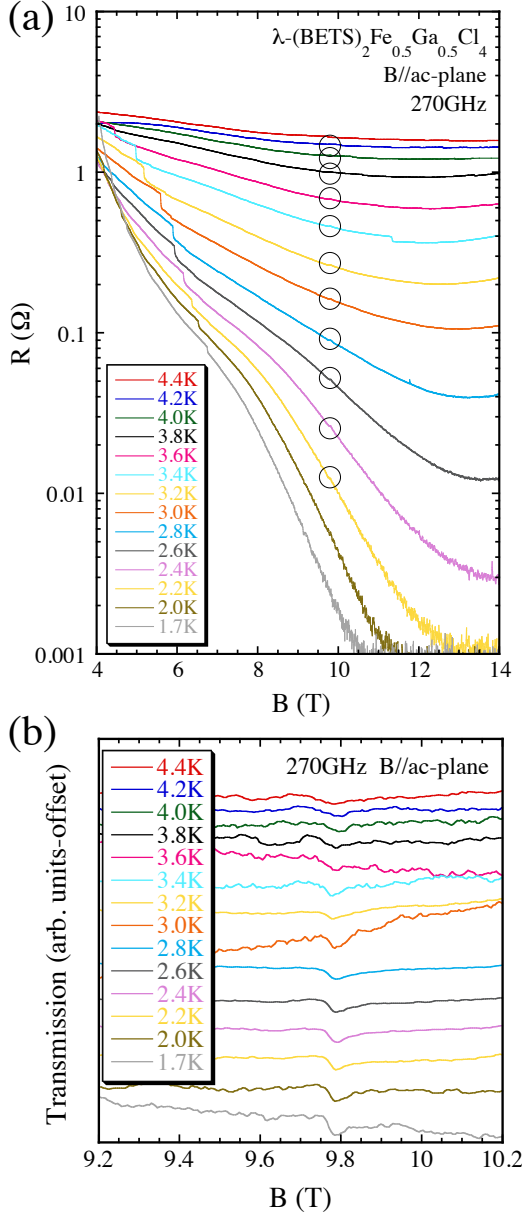


FIG. 6: The temperature dependence of (a) the magnetoresistance and (b) the ESR spectra for $x=0.5$ compound in the FISC phase ($B \parallel ac$ -plane). The open circles in (a) represent the observed resistance peaks at the resonance field.

phase at much lower field, show similar behavior. As shown in Fig. 8, the temperature dependence of the magnetoresistance exhibits a complicated behavior such as PM to FISC and SC to PM transitions, or the re-entrant PM-FISC-PM phase. However, the resistance anomalies at ~ 9.8 T where ESR transition occurs (open circle in Fig. 8) also appears at some intermediate temperature, i.e. from 2.0 to 2.8 K. Therefore, the behavior of resistance anomalies at the resonance field is common in λ -BETS system, and it only appears in the intermediate

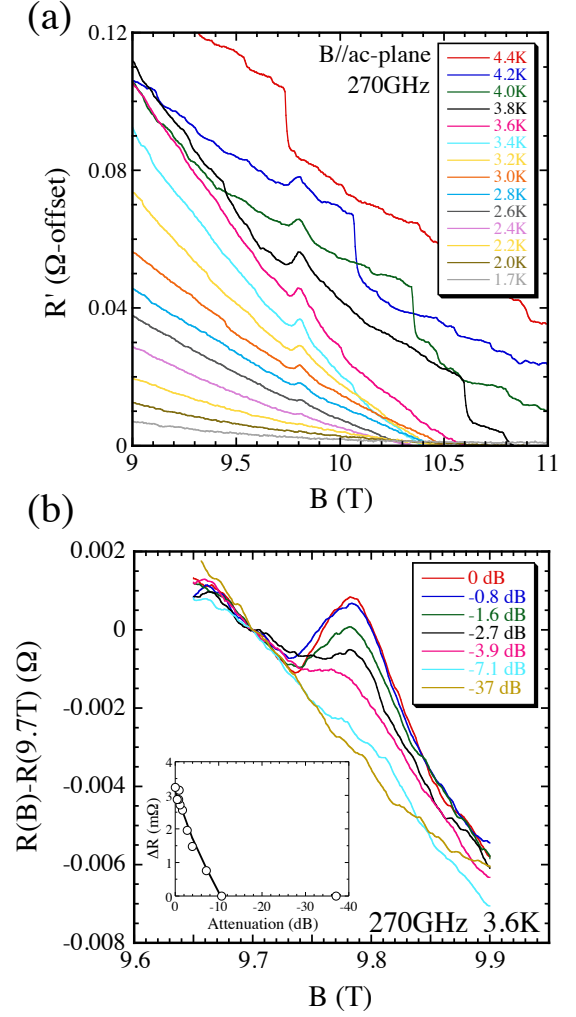


FIG. 7: (a) The temperature dependence of resistance peak for $x=0.5$ compound. The actual resistance is offset for each temperature for clarity. (b) The power dependence of the irradiated millimeter wave for 270 GHz at 3.6 K, which shows the maximum resistance peak ΔR . The inset is the attenuation diagram of ΔR .

temperature range.

IV. DISCUSSION

We have shown from the simultaneous transport and ESR measurements of the mixed compounds $x=0.34$, 0.5 and 0.6 that, coincident with the ESR resonance field, peaks in the magnetoresistance occur at intermediate temperatures within the FISC state.

In general, the observation of an ESR signal in a superconducting state is technically difficult due to the limit of the flux penetration depth. However, several ESR studies have been performed in the type-II superconductors which contains diluted magnetic impurities, and anoma-

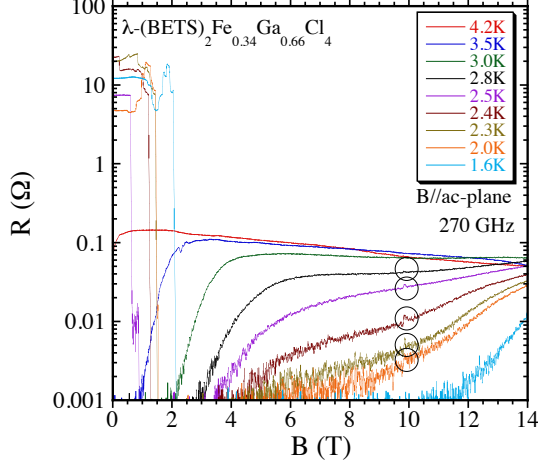


FIG. 8: The temperature dependence of the magnetoresistance for $x=0.34$ compound ($B\parallel ac$ -plane). The open circles represent the observed resistance peaks at the resonance field.

lies in the g -value, linewidth, and lineshape are observed below T_c .^{19,20} In contrast, for λ -(BETS)₂Fe _{x} Ga_{1- x} Cl₄, no drastic change was observed in the ESR signal of the FISC state. Since the linewidth is almost identical with one of the PM state, we conclude that the ESR signal observed in the FISC phase originates from the Fe³⁺ of the residual PM domains. This suggests that the FISC state in the λ -(BETS)₂Fe _{x} Ga_{1- x} Cl₄ system is an inhomogeneous state, and this is probably the reason why transport shows a smooth resistance change from the PM to FISC phase.

It is worthy to note that such inhomogeneous state might be due to disorder of the mixed compound system since the non-magnetic ions are introduced randomly to the sample. However, it is also possible that the inhomogeneous state is an intrinsic nature of the FISC state since smooth magnetoresistance change from PM to FISC state is also observed for the pure λ -(BETS)₂FeCl₄, especially in the temperature range above 0.5 K.³ Moreover, an anomalous broadening of the NMR line has been observed in the FISC phase for λ -(BETS)₂FeCl₄.¹² Although the authors explain this behavior by the existence of charge disproportionation, it is also possible that such inhomogeneous state is the origin of the NMR line broadening. We believe that spin fluctuation of Fe³⁺ ions plays an important role on the stability of the FISC state. When entering the FISC phase, the superconducting domains will gradually grow by increasing the magnetic field or decreasing the temperature since the fluctuations are suppressed. This is consistent with our temperature dependence of the ESR spectra, which suggest the reduction of the PM domains by decreasing the temperature. Therefore, spin fluctuation of Fe³⁺ ions is also one of the candidate for the origin of the inhomogeneous FISC state.

Next, we discuss the observed resistance anomaly at

the resonance field. When ESR transition occurs between the Zeeman splitting, there is always a thermal relaxation after the transition, which induces heat to the lattice. Since the resistance value in the FISC phase is temperature dependent due to the inhomogeneous nature, it is probable that this heat from the thermal relaxation increases the resistance at the resonance field. However, as shown in Figs. 4(a), 7(a) and 8, the resistance anomalies only appear in the intermediate temperature range. If the resistance anomaly is due to the thermal relaxation of ESR, this should be observed in any temperature range.

In addition, the maximum of the resistance peak, ΔR , is inversely proportional to the power attenuation as shown in the inset of Fig. 7(b). If we consider that the power absorbed by the spins heats the sample, and the heat leaks through the four gold wires, its relation can be expressed as follows,

$$P_{sample} = \frac{4S_{Au}}{l_{Au}} \int_{T_0}^{T_s} \kappa_{Au} dT. \quad (1)$$

P_{sample} is the power absorbed at the sample, T_s and T_0 are the sample's temperature and the base temperature, respectively. S_{Au} , l_{Au} , κ_{Au} and the factor are the area, length, thermal conductivity and number of gold wires, respectively. From this equation, one can easily notice that the temperature increase due to the thermal relaxation, $T_s - T_0$, should be proportional to P_{sample} . Hence, if the resistance peak is caused by the change of sample's temperature on resonance, ΔR should be linear with light power. However, what we see in the inset of Fig. 7(b) is that ΔR is linear to the power attenuation, which is linear in the log of the power, as mentioned in the previous section. Therefore, the thermal relaxation scenario can be excluded.

The alternative mechanism for the resistance anomalies is the FISC breakdown scenario by ESR. Due to the Jaccarino-Peter compensation effect, the effective field $\mu_B H_{eff}$ acting on the π -electrons is the sum of internal field created by the Fe³⁺ magnetic moments and the external field, i.e. $\mu_B H_{eff} = J_{\pi-d} S_d + \mu_B H_{ext}$ where $J_{\pi-d}$ is the π -d exchange interaction and S_d is the spin states of Fe³⁺.¹⁰ However, when the ESR transition occurs, the spin states changes from $S_d = -5/2$ to $-3/2$, especially at low temperature and high magnetic field. This corresponds to the spin flip of the Fe³⁺ spins and suggests that the angle and the absolute value of internal field change with the ESR transitions. This is crucial for the stability of FISC state which requires the magnetic field angle precision of a few degrees.³ Particularly when the magnetic field is near the boundary of the FISC phase, the compensation mechanism might be easily broken and the FISC state is destroyed. Therefore, the observed peaks in the resistance are possibly due to the breakdown of the FISC by ESR transitions.

Then, the question 'Why the peaks, which are expected to be related to the breakdown of the FISC state by ESR, are only observed in the intermediate temperature range?' arises. To answer, this behavior can be

explained from the inhomogeneous state, and the partial breakdown, of the FISC. As stated above, there are residual PM domains in the FISC phase since ESR is observed at low temperature. However, if the FISC domains are large enough so that there is a continuous superconducting path between the terminals, the resistance value remains minimal. Meanwhile, these FISC domains are diminished as a function of temperature, and PM domains start to appear within the conducting path. This behavior is confirmed by the resistance increase with temperature as shown in Figs. 4(a), 7(a) and 8. For an intermediate temperature range, there are some small FISC domains which break into the conducting path, and when such small domains are destroyed by the ESR, we are able to observe as a peak in the resistance. Finally, for higher temperature, tiny FISC domains are almost negligible in the resistance contribution, and therefore, the peak is no longer detected even if the ESR transition occurs. Therefore, we conclude that the resistance anomaly observed at the resonance field is due to the breakdown of the FISC triggered by the spin reversal of the Fe^{3+} spins located in the residual PM domains.

It is important to consider why the breakdown of the FISC state is very limited since ΔR of only few $\text{m}\Omega$ is observed. Firstly, the destroyed FISC state should rapidly recover due to the fast relaxation of Fe spins. The timescale of the AC resistance bridge cannot follow such rapid change in the resistance. Presumably, the observed resistance anomaly is a result of an averaged breakdown and recovering cycle of FISC domains. Secondly, due to the skindepth effect, the breakdown of the FISC only occurs nearby the surface of the sample. Since transport measures the resistivity of whole sample, we expect that the breakdown of FISC is very limited. Therefore, a very thin single crystal of about few μm is needed for the observation of larger breakdown in ΔR .

V. SUMMARY

In summary, we have performed simultaneous measurements of magnetotransport and ESR in the π -d molecular conductors λ -(BETS) $_2\text{Fe}_x\text{Ga}_{1-x}\text{Cl}_4$ ($x=0.34, 0.5$ and 0.6). By accurately applying the magnetic field parallel to the conducting plane, we have confirmed that ESR is observed even in the FISC state, which is consistent with our previous study.¹³ Meanwhile, the temperature dependence of our simultaneous measurements reveals an inhomogeneous FISC state down to 1.65 K.

We have also found that the ESR transitions of the Fe spins destroy the FISC state. This suggests that the FISC state can be controlled by radiation of electromagnetic wave, and superconducting to metal phase switching might be possible in the near future. Presently, due to the weak millimeter-wave power and the skin-depth effect, the destruction is partial where only a small change in the resistance is observed. In terms of ‘control’ of the superconductivity, millimeter-wave power that can saturate the ESR transition and very thin sample are needed, and this remains a subject for future investigation.

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