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Josephson vortex dynamics and Fulde-Ferrell-Larkin-Ovchinnikov superconductivity in the layered organic superconductor $\beta^{\prime\prime}$ -(BEDT-TTF)₂SF₅CH₂CF₂SO₃

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Phys. Rev. B **100**, 014515 — Published 17 July 2019

DOI: [10.1103/PhysRevB.100.014515](https://doi.org/10.1103/PhysRevB.100.014515)

**Josephson vortex dynamics and Fulde-Ferrell-Larkin-Ovchinnikov
superconductivity in layered organic superconductor**

β'' -(BEDT-TTF)₂SF₅CH₂CF₂SO₃

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(Dated: June 5, 2019)

Abstract

Interlayer resistance is reported in a layered organic superconductor β'' -(BEDT-TTF) $_2$ SF $_5$ CH $_2$ CF $_2$ SO $_3$ with $T_c \approx 5$ K. At 0.7 K in parallel magnetic fields, the interlayer resistance has nonzero values in a wide field region from 2 T to the upper critical field (~ 13 T), which can be ascribed to the Josephson vortex (JV) dynamics. Significant hysteresis between 2 T and 8 T, and a step-like structure between 4 T and 8 T are found, both of which become less evident with increasing temperature. In the second field-derivative curves, successive small dips are observed between 10 T and 12 T, which are ascribed to the commensurability (CM) effect between the JV lattice and the wave length λ_{FFLO} of the order parameter oscillation in the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase. The CM effect is observed only in nearly parallel fields, showing that the FFLO phase is stabilized only in a limited field angle region. The temperature-field phase diagram and λ_{FFLO} values are obtained.

I. INTRODUCTION

Layered organic superconductors have attracted much interest because of intriguing properties in high magnetic fields. Because of stacking structures of the organic superconducting (SC) and insulating layers [Fig. 1(a)], layered organic superconductors can be modeled as stacks of Josephson junctions. When the field is applied perpendicular to the layers, the flux lines penetrate the SC layers and form vortices. This vortex formation (orbital effect) strongly destabilizes the superconductivity, leading to a small upper critical field (H_{c2}). By contrast, when the field is applied parallel to the layers, most of the flux lines can penetrate between the SC layers, and then the orbital effect is strongly quenched. This is the main reason why H_{c2} in the parallel direction is much higher than that in the perpendicular direction. In addition, the superconductivity is in a clean limit for most of organic superconductors. The two conditions **discussed** above, the orbital effect is quenched and the superconductivity is in the clean limit, are particularly advantageous to the emergence of a novel SC phase, the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase.[1, 2] At sufficiently below the critical temperature T_c and in parallel fields, the FFLO superconductivity can survive even above the Pauli paramagnetic limit H_{Pauli} , which is determined by the Zeeman effect.[3] In the FFLO phase, the Cooper pairs are formed between the up and down spins on the polarized Fermi surfaces and, consequently, have a non-zero center-of-mass momentum

\mathbf{q} . The finite \mathbf{q} value leads to an order parameter oscillation (periodic nodal lines) in the real space, $\Delta = \Delta_0 \cos(\mathbf{q}\mathbf{r})$. [1, 2] It is expected that the FFLO transition occurs at $\sim H_{\text{Pauli}}$. In layered organic superconductors, a FFLO phase transition in the superconducting phase was first suggested by a tuned-circuit differential susceptometer experiment for κ -(BEDT-TTF)₂Cu(NCS)₂, [4] and then its transition has been observed in various measurements. [5–8] After that, a FFLO phase transition has been found in λ -(BETS)₂X (X = GaCl₄, FeCl₄), [9–15] β'' -SF₅, [16–19] and β'' -(BEDT-TTF)₄[(H₃O)Ga(C₂O₄)₃]C₆H₅NO₂. [20]

In highly two dimensional (2D) superconductors, magnetic flux lines can be decomposed into two parts, pancake vortices (PVs) in the SC layers and Josephson vortices (JVs) in the insulating layers between the SC layers. In contrast to PVs, JVs have no normal cores, and thus JVs are pinned very weakly: JVs are easily driven by a perpendicular current. If the JVs are driven, nonzero interlayer resistance is observed even in the SC phase. At sufficiently low temperatures, JVs will form a lattice in the insulating layers. The JV lattice constant l decreases with increasing field. In the FFLO phase, the \mathbf{q} value increases with field, [21] giving the wavelength of the order parameter oscillation $\lambda_{\text{FFLO}} = 2\pi/q$. Figure 1(b) presents a schematic picture of the JV lattice and order parameter oscillation. Both the length scales, l and λ_{FFLO} decrease with increasing field, and thus l becomes commensurate with λ_{FFLO} at certain fields but incommensurate at the other fields. In the commensurate case, the JV lattice will be relatively strongly pinned by the periodic structure of the order parameter, giving a relatively small resistance. If the commensurate condition is satisfied at some different fields, we could observe periodic oscillatory behavior of the interlayer resistance as the field increases, which is called commensurability (CM) effect. The CM effect was first theoretically predicted by Bulaevskii et al. [22] Although FFLO phases have been investigated in various layered organic superconductors, the CM effect is experimentally observed only in the FFLO phases of λ -(BETS)₂FeCl₄ [13] and β'' -(BEDT-TTF)₄[(H₃O)Ga(C₂O₄)₃]C₆H₅NO₂. [20] When the field is tilted from the layers, the oscillatory behavior disappears, which shows that the FFLO phase is easily broken by the orbital effect. From the CM conditions, λ_{FFLO} is estimated to range from 20 nm to 150 nm.

The SC state in β'' -(BEDT-TTF)₂SF₅CH₂CF₂SO₃ (hereafter, β'' -SF₅ salt) is expected to be even more 2D than the above organic superconductors and extensive efforts have been devoted to discovering the FFLO phase transition. [16–19] In a low temperature region, an anomaly due to the FFLO phase transition at 9.3–10.5 T has been reported in the RF re-

sponse (resonance frequency change of a tunnel diode oscillator),[16] NMR,[18] and magnetic torque measurements.[19] However, no convincing evidence of the FFLO transition has been obtained in the heat capacity because of the existence of the vortex phase transitions.[17] Recent magnetocaloric effect (MCE) measurements, which are very sensitive to the magnetic entropy change, clarified the presence of two anomalies, associated with hysteresis and latent heat, below H_{c2} . [19] The results show that a FFLO phase transition takes place at ~ 9.5 T and then a melting transition of the JVs at a higher field. The result is a direct evidence showing that the FFLO and JV melting transitions take place independently. Such successive phase transitions will lead to characteristic vortex dynamics, which is selectively observed in the interlayer resistance. To further investigate the vortex dynamics, we have performed the interlayer resistance measurements for the β'' -SF₅ salt. We observe peculiar behavior in the field dependence in a wide field region below H_{c2} . We discuss the behavior in terms of the JV dynamics, which strongly depends on the JV and FFLO phases.

II. EXPERIMENTS

Single crystals of β'' -(BEDT-TTF)₂SF₅CH₂CF₂SO₃ were synthesized by a standard electrochemical method.[23] The crystal used for these measurements is obtained from the same batch reported in our previous studies.[19] Two gold wires of 10 μ m in diameter were attached on both sides of a single crystal (conducting *ab* plane) by carbon paste. The interlayer resistance was measured by a conventional four-probe AC technique with a frequency of ~ 15 Hz. The electric current I with a density of 1.5 mA/cm² is applied perpendicular to the conducting layers. The schematic of the crystal is depicted in the inset of Fig. 2(a). The crystals were slowly cooled down to 0.5 K at a rate of 1 K/min using a ³He refrigerator with a 17 T superconducting magnet.

III. RESULTS

Figure 2(a) presents the field dependence of the interlayer resistance in perpendicular fields for $T = 1.5$ K. The resistance rapidly increases above 0.3 T, has a sharp peak at 0.6 T and then decreases. A similar peak is found in other organic superconductors.[20, 27–30] Above ~ 8 T, where the resistance gradually increases with field, we observe Shubnikov-

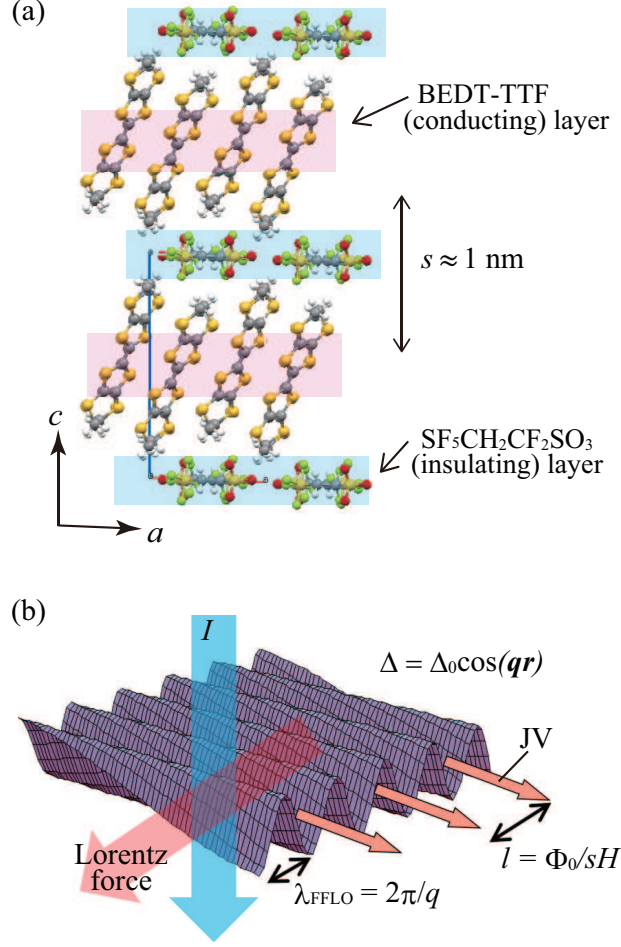


FIG. 1. (color online) (a) Schematic picture of the crystal structure of β'' -SF₅ salt with the space group $P\bar{1}$; $a = 0.91536$ nm, $b = 1.14395$ nm, $c = 1.74905$ nm, $\alpha = 94.316^\circ$, $\beta = 91.129^\circ$, $\gamma = 102.764^\circ$. [23] This salt is composed of BEDT-TTF layers and SF₅CH₂CF₂SO₃ layers. The layer spacing s is indicated. (b) Schematic picture of superconducting order parameter $\Delta(\mathbf{r})$ and JVs in the FFLO phase. The center-of-mass momentum of the Cooper pair \mathbf{q} gives the wavelength of the order parameter oscillation λ_{FFLO} . JVs, which are formed in the insulating layers with a lattice constant l , can be driven by a perpendicular current I .

de Haas (SdH) oscillations, whose Fourier transform spectrum is shown in the inset. The frequency $F = 200$ T, corresponding to $\sim 5.8\%$ of the first Brillouin zone, is consistent with the previous report. [24, 31, 32] At higher temperatures, the resistance peak and SdH oscillations are reduced. The observation of the SdH oscillations clearly indicates that the superconductivity is in the clean limit. The Fermi surface calculated by a tight binding approximation [23] is presented in Fig. 2(b). There are a pair of 1D and a 2D Fermi surfaces.

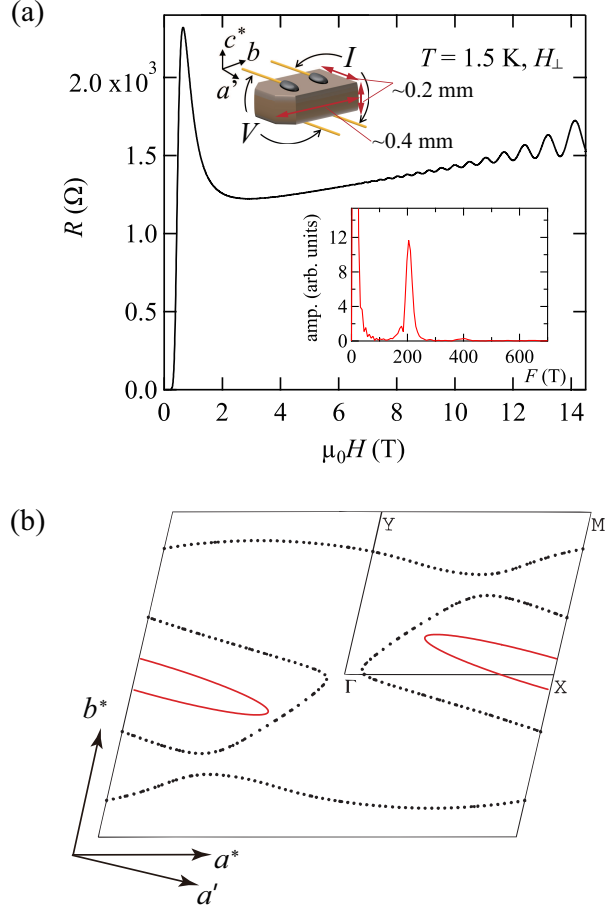


FIG. 2. (color online) (a) Interlayer resistance in perpendicular magnetic fields at 1.5 K. Inset: Fourier transform spectrum of the SdH oscillation, and a schematic of the single crystal and the electric contact configuration. (b) Fermi surface calculated by tight binding approximation (black dots).[23] For comparison, the 2D Fermi surface determined from AMRO measurements is shown by a red curve.[24]

In the angular dependent magnetoresistance oscillation (AMRO) measurements,[24, 27] a closed orbit (red curve) is obtained, which is much smaller and more elongated roughly along a^* than the calculation [Fig. 2(b)].

Figure 3(a) shows typical field dependences of the interlayer resistances and their second derivative curves at 0.7 K. **The magnetic field is applied nearly along the a' axis, but parallel to the SC layers within the accuracy of 0.1° .** The dotted and solid curves present the results for the up and down sweeps, respectively. The resistance increases with field above 2 T, has a step-like structure between 4 T and 10 T, and then steeply increases again. Since H_{c2} is

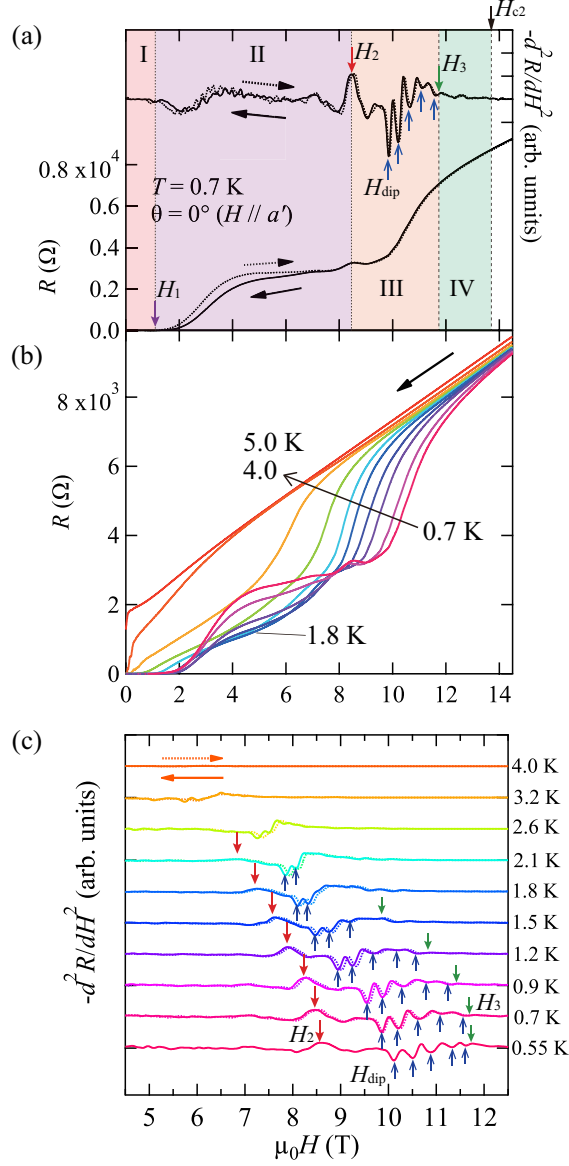


FIG. 3. (color online) (a) Magnetic field dependence of interlayer resistance and second derivative curve at 0.7 K. The H_{c2} value was determined by the TE[25] and RF response[16] measurements. (b) Interlayer resistance at various temperatures, and (c) second derivative curves of the resistance for the up and down sweeps. H_{dip} is not evident above 2.1 K. The characteristic fields H_1 - H_3 , and dip fields H_{dip} are indicated by arrows.

~ 13 T at 0.7 K,[16, 19] the non-zero value of the resistance in the wide field region down to 2 T. We observe significant hysteresis between 2 T and 8 T. The presence of the hysteresis shows that the JV dynamics leading to energy dissipation is different between the up- and

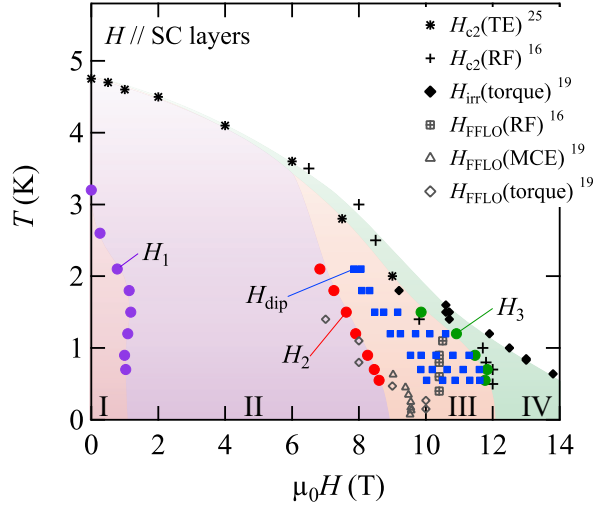


FIG. 4. (color online) Magnetic phase diagram for $H \parallel \text{SC layers}$. The characteristic fields, H_1 - H_3 and H_{dip} are indicated by color symbols. For comparison, H_{c2} determined by TE and RF, irreversibility field (H_{irr}) by torque, and H_{FFLO} by RF, MCE, and torque measurements are also indicated by black symbols. The SC phase is divided into four regions: I) $H < H_1$, II) $H_1 < H < H_2$, III) $H_2 < H < H_3$ and IV) $H_3 < H < H_{c2}$.

down-sweeps in this field region, more JVs are driven (pinned) for the up (down)-sweep. The fact may suggest the presence of metastable JV configurations, depending on the pinning site distributions.

First, we define H_1 , where the finite resistance appears. The H_1 is roughly defined as the **depinning** field of the JVs. At ~ 8.5 T (H_2), a peak is seen in the second derivative curve, below which the hysteresis is evident. In the range between ~ 9.5 T and ~ 12 T, oscillatory behavior is observed, whose dip fields are denoted as H_{dip} . We note that the oscillatory behavior is reproduced between the up and down sweeps. At ~ 12 T (H_3), we see a small peak. Above H_3 , no significant structure is seen; the second derivative curve becomes almost flat. In previous studies, the FFLO transition is found at ~ 9.5 T below 1.5 K.[16, 18, 19] Therefore, the oscillatory behavior will be ascribed to the CM effect. We have measured the resistance at lower currents down to $\sim 30 \mu\text{A}/\text{cm}^2$, but no appreciable current dependence of H_{dip} is observed.

As the temperature increases, the finite resistance appears down to a lower field region [Fig. 3(b)], showing that the JVs are driven more easily by the perpendicular current.

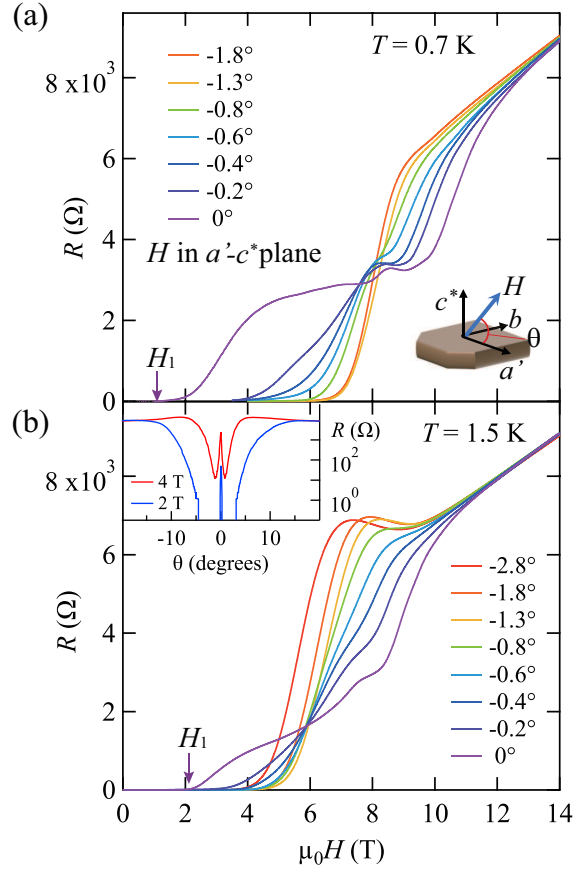


FIG. 5. (color online) Magnetic field dependence of the interlayer resistance at various field angles (a) at 0.7 K and (b) at 1.5 K. Inset in (b): Semilog plot of interlayer resistance as a function of the field angle θ at 1.5 K for $\mu_0 H = 2$ and 4 T.

The step-like structure between 4 T and 10 T is smeared with increasing temperature. The resistance in this region decreases with increasing temperature up to 2.1 K and then increases again. This behavior clearly shows that the JVs are more strongly pinned up to 2.1 K. This seems very anomalous behavior, whose origin is not clear at present. At 5 K, the resistance has a steep increase above 0.1 T and then linearly increases with increasing field. This linear dependence is typical behavior for 2D electronic states.[33] As seen in the $R(H)$ and derivative curves [Fig. 3(c)], H_2 , H_3 and H_{dip} gradually decrease with increasing temperature. The peak at H_2 and oscillatory behavior are evident up to ~ 2.1 K. The peak at H_3 is slightly visible up to 1.5 K.

From the above results, we can draw a phase diagram as presented in Fig. 4. The H_1

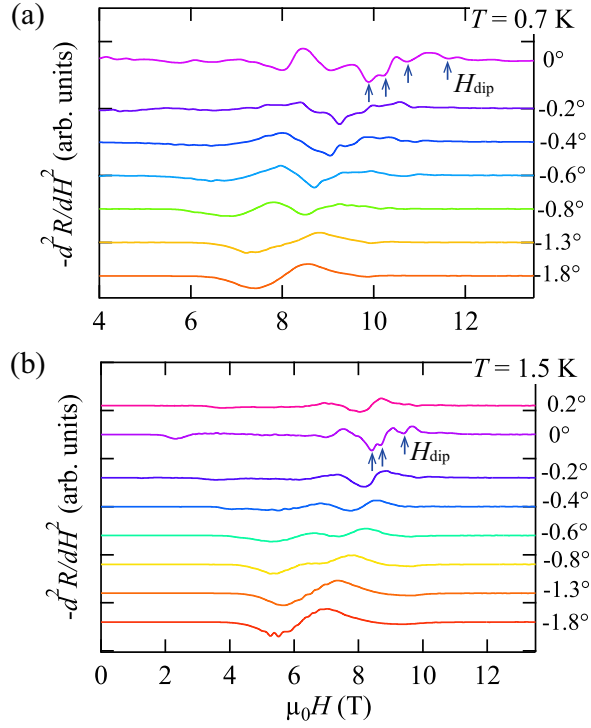


FIG. 6. (color online) Second derivative curves $-d^2R/dH^2$ at various field angles (a) at 0.7 K and (b) at 1.5 K. The curves are shifted for clarity. As the field is tilted from $\theta = 0^\circ$ ($H \parallel$ SC layers), the dips arising from the CM effect indicated by blue arrows are suppressed and disappear.

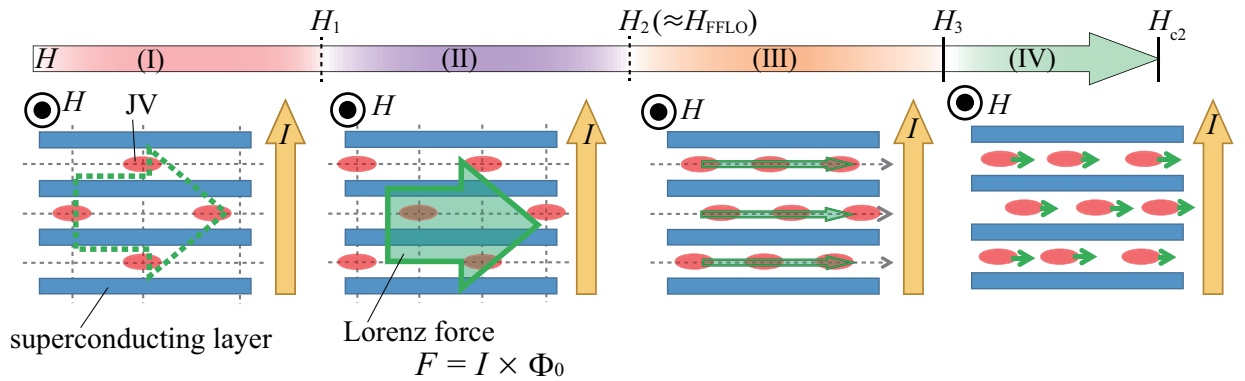


FIG. 7. (color online) Schematic picture of the JV dynamics. The SC phase could be divided into four regions: I) the JV lattice is strongly pinned, II) the JV lattice is collectively driven by the current, III) the JV layers are decoupled and the JV lattice in each layer is driven independently, and IV) the JV lattice melts.

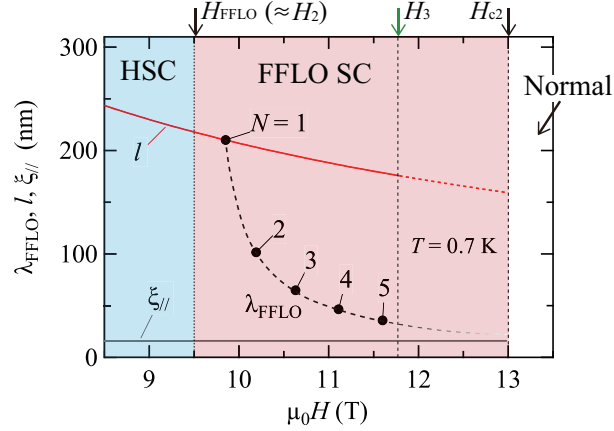


FIG. 8. (color online) The estimated wave length of the FFLO order parameter oscillation, λ_{FFLO} at 0.7 K (black circles). The black dashed curve shows a guide to the eye. The calculated JV lattice constant l is shown by red curve. The JV lattice constant l is expected to have discrete values, depending on the boundary condition at the sample edges.[26] For simplicity, the field dependence of l is indicated by a continuous curve. The FFLO phase transition takes place at ~ 9.5 T (H_{FFLO}). Above H_3 , where the JV lattice melts, l is not defined and thus the CM effect is not observed.

value is almost constant up to ~ 1.8 K and then decreases with increasing temperature. The H_{c2} values have been determined by thermal expansion (TE),[25] and RF response measurements,[16] which roughly correspond to the irreversibility field H_{irr} determined by the magnetic torque.[19] The FFLO phase transition field H_{FFLO} obtained by the magnetic torque measurement approximately corresponds to H_2 . The H_{FFLO} value by the MCE measurements [19] smoothly connects to H_2 . Although H_{FFLO} determined by the RF response [16] disagrees with them, our results consistently show that the regions III and IV correspond to the FFLO phase. The FFLO phase seems to survive up to ~ 3 K at ~ 8 T, which can be defined as the tricritical point.

Figures 5(a) and 5(b) present the resistance at various field directions at 0.7 K and 1.5 K, respectively. Here, we also define H_1 , where the finite resistance appears. The H_1 value has a minimum for $\theta = 0^\circ$ and increases as the field is tilted. As the field is further tilted, H_1 decreases. This behavior is evident in the angular dependence of the resistance as shown in the inset of Fig. 5(b). We see a sharp peak at $\theta = 0^\circ$, which is ascribed to the JV dynamics. As the field is tilted, the flux lines penetrate the SC layers, where they are strongly pinned. This strong pinning reduces the resistance. As the field is further tilted, the resistance

increases due to less pinning (reduction of the order parameter).

The step-like structure between 4 T and 10 T at 0.7 K [Fig. 5(a)] is quickly smeared out as the field is tilted. At 1.5 K [Fig. 5(b)], the step-like structure is not evident even at $\theta = 0^\circ$. As the field is tilted, a broad bump appears near H_{c2} for $|\theta| > 0.8^\circ$. The second derivative curves are presented in Figs. 6(a) and 6(b). We note that the CM effect (H_{dip}) is observable only in a limited field angle region for $|\theta| > 0.4^\circ$ (0.2°) at 0.7 K (1.5 K). The results show that the FFLO phase is stable in a very small angle region, consistent with previous reports.[13, 20]

IV. DISCUSSION

A. Field dependence of JV dynamics

The non-zero resistance in the wide field region below H_{c2} shows that JVs are easily driven by the perpendicular current. From the field dependence in Fig. 3(a), the vortex dynamics will be divided into four regions: I) $H < H_1$, II) $H_1 < H < H_2$, III) $H_2 < H < H_3$ and IV) $H_3 < H < H_{c2}$ (Fig. 7). In region I, we observe zero resistance. At sufficiently low temperatures and fields, the SC layers are strongly Josephson-coupled, the JVs form a 3D lattice and will be collectively pinned (not driven by the current). In region II, the step-like structure is seen. The nonzero resistance shows that the JV lattice is deppined and collectively driven by the current. **As the field increases, the interlayer coupling of the JVs weakens (region III). Therefore, the JV layers could be decoupled by the perpendicular current although the JVs form a 3D lattice in zero current. We expect that the JV lattice in each layer is independently driven by the perpendicular current. This decoupling, which effectively reduces the pinning force, will lead to an increase of the interlayer resistance.** As the field further increases, the JV lattice will melt as has been observed in the MCE.[19] The region IV will be ascribed to the JV melting region. The JV melting is consistent with the absence of any structure in the second derivative curves. Although the FFLO phase should be stable up to H_{c2} ,[19] it will be impossible to observe the CM effect in the melting phase of the JVs. In this melting region, the resistance continuously reaches a normal state value with increasing field. Here we should note that the thermodynamic phase transition of the JVs is only the melting transition at H_3 .

TABLE I. Superconducting parameters in layered organic superconductors showing CM effect

material	T_c [K]	$\mu_0 H_{c2\perp}$ [T]	$\mu_0 H_{c2\parallel}$ [T]	$H_{c2\parallel}/H_{c2\perp}$	$\mu_0 H_{\text{FFLO}}$ [T]	$\mu_0 H_{\text{FFLO}}/T_c$ [T/K]	Ref.
λ -(BETS) $_2$ FeCl $_4$	3~4	-	16*	-	7*	1.8~2.1	[13]
β'' -Ga	4.8	2.5	21	8.4	15	3.1	[20]
β'' -SF $_5$	4.7~5.2	1.3	14	11.5	9.5-10	1.9~2.1	this study

*: estimated by considering the internal field.[13]

β'' -Ga: β'' - (BEDT-TTF) $_4$ [(H $_3$ O)Ga(C $_2$ O $_4$) $_3$] C $_6$ H $_5$ NO $_2$

Ref.: references

B. CM effect and field dependence of λ_{FFLO}

The dips of the $-d^2R/dH^2$ curves in the FFLO phase [Figs. 3(c), 6(a) and 6(b)] can be ascribed to the CM effect between λ_{FFLO} and l . In the FFLO phase, the Josephson coupling strength (Josephson current) between the layers periodically oscillates, due to the order parameter oscillation. This situation will form a periodic potential for the JVs [Fig. 1(b)], which is the main origin of the CM effect. The JV lattice configuration is theoretically discussed based on Lawrence-Doniach model by Nonomura and Hu.[26] According to this study, the JVs have complicated periodic structures, which are characterized by two lattice constants, l (parallel to layer) and l' (perpendicular to layer). The structures depend on a normalized field given by $h = 2\pi\gamma s^2 H/\Phi_0$ and the order parameter amplitude. Here Φ_0 is the flux quantum and γ is the anisotropy ratio, which is estimated to be 400-800.[34] For $h \gtrsim 1.1$ ($\mu_0 H \gtrsim 1$ T), the JVs enter each layer equally ($l' = s$), forming a same lattice constant l in each layer. The JV lattice constant is given by $l = \Phi_0/sH$, where s is the layer spacing ($s \approx 1$ nm). In the homogeneous superconducting (HSC) phase, λ_{FFLO} is infinite ($\mathbf{q} = 0$). However, with increasing magnetic field, λ_{FFLO} is expected to jump to a finite value at the first order FFLO phase transition. After that, λ_{FFLO} will decrease with increasing field up to H_{c2} . Since the stability of the FFLO phase is closely related to the nesting instability of the Fermi surface, the optimum \mathbf{q} vector will depend on the detailed structure of the anisotropic Fermi surfaces [Fig. 2(b)]. **Since most parts of the Fermi surfaces are parallel to the a' axis, the optimal FFLO \mathbf{q} vector will be nearly along b^* axis.** For simplicity, we estimated λ_{FFLO} (Fig. 8) on the basis of the following assumptions: (i) \mathbf{q} is parallel to k_b ($\mathbf{q} \perp \mathbf{H}$) (ii) l/λ_{FFLO} is given by simple integers N ; $l/\lambda_{\text{FFLO}} = N$, (iii) λ_{FFLO} is an order of

the in-plane coherence length ξ_{\parallel} (~ 16 nm) near H_{c2} , (iv) N is close to unity at H_{FFLO} . The estimated value is shown by black circles in Fig. 8. We note that λ_{FFLO} rapidly decreases with increasing field above H_{FFLO} and becomes comparable to ξ_{\parallel} at H_{c2} . A similar upward curvature has been obtained for other organic superconductors.[13, 20]

The \mathbf{q} value will approximately correspond to the wave number difference between the up and down spin Fermi surfaces, polarized by the Zeeman effect. Assuming $\mathbf{q} \perp \mathbf{H}$, we estimate the \mathbf{q} value from the relation, $\mathbf{q} = g\mu_B \mathbf{H} \mathbf{k}_F / 2E_F$, where g is the g -value (~ 2), \mathbf{k}_F ($|| \mathbf{q}$) is the Fermi wave number and E_F is the Fermi energy. We obtain $E_F = \hbar^2 A_F / 2m^* \pi \approx 130$ K, where A_F is the cross section of the 2D Fermi surface and $m^* (\approx 2m_0)$ is the effective mass.[31, 32, 35] The above relation gives $q \approx 4.0 \times 10^7 \text{ m}^{-1}$ at 10 T, corresponding to $\lambda_{\text{FFLO}}^{\text{cal}} \approx 160$ nm. This value is reasonably consistent with $\lambda_{\text{FFLO}} \approx 210$ nm determined by the dip fields.

Tachiki et al. theoretically discussed the superconducting order parameters of a FFLO phase and obtained λ_{FFLO} as a function of the magnetic field, which included an orbital effect.[21] They show that the $\lambda_{\text{FFLO}}(H)$ curve has an upward curvature with decreasing field, and it ranges from $30\xi_{\parallel}$ to $13\xi_{\parallel}$. In the absence of the orbital effect, the FFLO phase will be stable in a higher field region, giving a shorter λ_{FFLO} . Shimahara theoretically estimated λ_{FFLO} for a 2D system having a simple Fermi surface and showed that λ_{FFLO} at H_{c2} decreases down to $\pi\xi_{\parallel}$ with decreasing temperature.[36] In the β'' -SF₅ salt, we obtain $\lambda_{\text{FFLO}}/\xi_{\parallel} = 2.2$ -13.1, consistent with the theories. We note that H_{dip} decreases with increasing temperature in Fig. 4: $H_{\text{dip}} \approx 10$ T at 0.55 K and $H_{\text{dip}} \approx 8$ T at 2.1 K for $N = 1$. The result suggests that λ_{FFLO} at H_{c2} increases with increasing temperature. The behavior is probably consistent with the theoretical prediction, λ_{FFLO} on the H_{c2} curve **diverges at** the tricritical point.[36] The above estimation depends on the angle between \mathbf{q} and \mathbf{H} . For instance, when the \mathbf{q} vector is tilted by θ from the field direction, λ_{FFLO} becomes smaller by a factor $\cos(\theta)$. At present, we have this ambiguity in the estimation. At sufficiently low temperatures, FFLO phases with multi \mathbf{q} vectors are theoretically expected to appear.[37] It will lead to successive first order FFLO transitions. So far, no evidence of such multiphase transitions has been seen.

Among various layered organic superconductors, the CM effect has been observed for λ -(BETS)₂FeCl₄[13] and β'' -(BEDT-TTF)₄[(H₃O)Ga(C₂O₄)₃]C₆H₅NO₂. [20] **The CM effect will become evident when the pinning potential of the JVs by sample inhomogeneity (defects**

or impurities) is comparable to that by the order parameter oscillation. It means that sufficiently high two dimensionality and quality of the samples are required for the observation of the CM effect. The $H_{c2\parallel}/H_{c2\perp}$ value is a good measure of the two dimensionality of the superconductivity. Table I presents the SC parameters for layered organic superconductors showing the CM effect in the FFLO phase. We note that the β'' -SF₅ salt has a highest value of $H_{c2\parallel}/H_{c2\perp}$. The fact shows that the JVs are less pinned, which will be the main reason of the finite resistance in the wide field region.

V. CONCLUSIONS

We report systematic measurements of the interlayer resistance in the highly 2D layered organic superconductor, β'' -(BEDT-TTF)₂SF₅CH₂CF₂SO₃ with $T_c \approx 5$ K. At 0.7 K in parallel magnetic fields, the JV dynamics are divided into four regions: I) the JV lattice is strongly pinned, II) the JV lattice is collectively driven by the current, III) the JV layers are decoupled and driven independently, and IV) the JV lattice melts. In region II, we observe large hysteresis in the field dependence, suggesting the presence of metastable vortex configuration. In the $-d^2R/dH^2$ curves, we observe successive dips between 10 T and 12 T (region III), which are ascribed to the CM effect between the JV lattice and the order parameter oscillation of the FFLO phase. In region IV, we do not observe the CM effect because of the melting of the JVs. The CM effect is observable only in a limited field angle region for $|\theta| > 0.4^\circ$ (0.2°) at 0.7 K (1.5 K); the FFLO phase is stable in a very small angle region. On the reasonable assumptions, we estimate the field dependence of λ_{FFLO} , which ranges from $\sim 2.2\xi_{\parallel}$ at $\sim H_3$ to $\sim 13.1 \xi_{\parallel}$ at $\sim H_{\text{FFLO}}$.

ACKNOWLEDGMENTS

This work was supported by a Grant-in-Aid for Scientific Research from MEXT, Japan (No. 17H01144). Work at ANL was supported by U. Chicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. J. A. S. acknowledges support from the Independent Research/Development program while serving

at the National Science Foundation.

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