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Anisotropic magnetoresistance in the organic superconductor β"–(BEDT-TTF)2SF5CH2CF2SO3

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Anisotropic magnetoresistance in the organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$

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In this paper, we report transport measurements of interlayer magnetoresistance with field parallel and perpendicular to the current direction in an all organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. For $H\|I$, the isothermal magnetoresistance $R(H)$ at low temperatures ($T\leqslant T_c$) displays a peak effect as a function of field. For $H\perp I$, $R(H)$ increases monotonically with increasing field. The results are very analogous to the interlayer magnetoresistance in κ-(BEDT-TTF)$_2X$ compounds. The observation of the peak effect or negative magnetoresistance in different systems for $H\|I\perp$ plane suggests that it is intrinsic to the layered organic superconductors. For $H\perp I$, the large positive magnetoresistance is in a general agreement with a two band model for charge transport. [S0163-1829(99)07405-6]

I. INTRODUCTION

Interlayer transport in layered systems has been of recent interest.[1–7] In anisotropic cuprates such as $Bi_2Sr_2CaCu_2O_{8+x}$, interlayer resistivity exhibits a semiconducting temperature dependence, while the in-plane resistivity is metallic. With applied field perpendicular to the superconducting layers, the semiconducting behavior is pushed to a lower temperature.[4,6] In layered organic superconductors, especially the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br and κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ salts, interlayer resistance is typically three orders of magnitude larger than the in-plane resistivity.[8,9] However, the temperature dependence is qualitatively similar, i.e., semiconducting for temperature above about 100 K and metallic for temperature below it. Furthermore, interlayer transport in these materials has shown interesting field and temperature-dependent magnetoresistance peak effect.[10–14] Various models including vortex-lattice interaction,[10] presence of magnetic impurities,[11] stacked Josephson-junction model with a field dependent quasiparticle tunneling[12–14] have been proposed to explain the peak effect, however, the origin remains controversial.[15]

To understand the mechanism in the interlayer charge transport, we have performed transport measurement on a highly two-dimensional all organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$.[16,19,20] The β'' structure contains layers of nearly parallel BEDT-TTF molecules, which are canted with respect to the stacking axis. In contrast the κ-type structures contain orthogonal BEDT-TTF molecules. Unlike the κ-(BEDT-TTF)$_2X$, where resistivity at ambient pressure shows a broad peak at near 100 K, both the in-plane and interlayer resistivity are metallic from room temperature down. The superconducting transition temperature is about 5 K, considerably smaller than 11 and 10 K for the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br and κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ salts, respectively. A comparative study with the κ-phase superconductors is thus highly desirable.

In this paper, we report measurements of interlayer magnetoresistance with field parallel and perpendicular to the current direction in the organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. For $H\|I$, the isothermal magnetoresistance $R(H)$ displays a peak effect as a function of field. For $H\perp I$, $R(H)$ increases monotonically with increasing field. The results are very analogous to the interlayer magnetoresistance in κ-(BEDT-TTF)$_2X$ compounds.[10–14] The similarity for the two different systems suggests that the peak magnetoresistance for $H\|I$ is intrinsic to the layered structure of the organic superconductors. For $H\perp I$, the magnetoresistance is in a general agreement with a two band model for charge transport.

II. EXPERIMENT

Single crystals of β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ were synthesized by the electrocrystallization technique described elsewhere.[16] Several crystals were used in these measurements with average dimensions of $1\times0.78\times0.33$ mm. Extensive measurements were made on one crystal with $T_c\sim5$ K. The T_c is defined as the midpoint in the resistive transition. Depending on the cooling rate, T_c can be varied from ~5 to 5.5 K. The room-temperature interlayer resistivity is about 700 Ω cm and the in-plane resistivity about 0.2 Ω cm. The resistivity ratio between room temperature and superconducting transition temperature is $[\rho_\perp(300\,\mathrm{K})/\rho_\perp(6\,\mathrm{K})]\sim230$. The interlayer resistance was measured with use of the four-probe technique. Contact of the gold wires to the sample was made with a Dupont conducting paste. Typical contact resistances between the gold wire and the sample were about 10 Ω. A current of 1 μA was used to ensure linear I-V characteristics. The voltage was detected with a lock-in amplifier at low frequencies of about

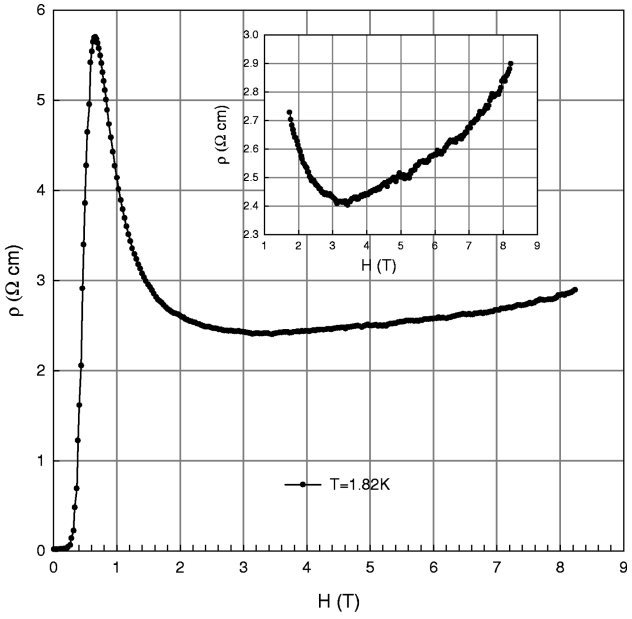

FIG. 1. Interlayer resistivity as a function of field for $H\perp$ plane at $T=1.82$ K. The inset is an expanded view of $\rho(H)$ at high fields.

312 Hz. The samples were cooled slowly to below the superconducting transition temperature with the field parallel and perpendicular to the crystallographic b axis. To avoid pressure effect due to solidification of grease, the sample was mechanically held by thin gold wires. Because of the small sample size, a misalignment of up to 5 degrees was possible.

III. RESULTS AND DISCUSSIONS

Shown in Fig. 1 is a typical plot of the interlayer resistivity as a function of field at $T=1.82$ K. The midpoint of resistive transition is about 5.5 K for the sample studied. Clearly, the resistivity ρ starts to rise rapidly at an onset field of about 0.2 T. ρ reaches a peak value at a peak field $H_{peak}\sim 0.7$ T. For field greater than H_{peak}, the resistivity decreases sharply with increasing field. At about 3 T, the slope dR/dH changes sign and becomes small and positive, as shown in the inset. If we compare the values of resistivity at 3 T and at the peak, it is easy to see $[\rho_{peak}/\rho(3\text{ T})]>2$.

Figure 2 shows the overlay of the resistivity peak as a function of field at various temperatures, $T=1.82$, 2, 2.2, 2.6, 3, 3.5, 4, 4.5, and 5 K. With increasing temperature, the peak shifts toward zero field and the magnitude of the peak increases initially and reaches a maximum at around 2.6 K. It decreases with further increase in temperature. Plotted in the inset is the peak field versus the temperature. H_{peak} increases monotonically with decreasing T. The solid line is a fit to $H_{peak}=H_o(1-T/T_c)^n$, with $H_o=1.5\pm 0.2$ T, $n=2.1\pm 0.1$, $T_c=5.4\pm 0.2$ K. The temperature dependence of the peak field demonstrates clearly that the peak effect is associated with the superconductivity, since it disappears above T_c. The temperature dependence of $H_{peak}(T)$ in the temperature range investigated is reminiscent of the temperature dependence of the critical fields in the cuprate superconductors. The characteristic field where the magnetoresistance is again positive decreases with increasing temperature. For temperature above T_c, magnetoresistance is always small and positive.

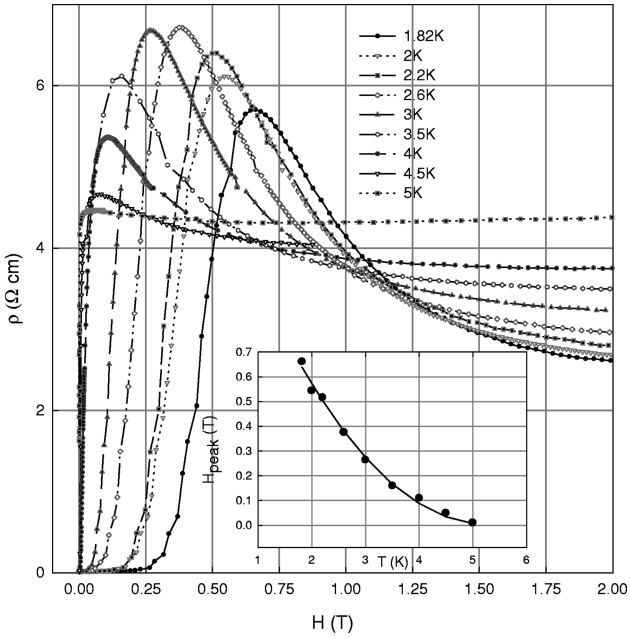

FIG. 2. Interlayer resistivity peak as a function of field for $H\perp$ plane at various temperatures. The inset is a plot of peak field versus temperature and the line is a fit.

Plotted in Fig. 3 is an overlay of interlayer resistivity at $H=0$ and 2 T, in comparison with $\rho_{peak}(T)$. At $H=2$ T, the resistivity decreases monotonically with decreasing temperature. The offset for $\rho(H=2\text{ T})$ above T_c from the zero-field data is due to the positive magnetoresistance. At lower temperatures, the $\rho_{peak}(T)$ is clearly displaced from the $\rho(H=2\text{ T})$ curve, and reaches a maximum at around 2.6 K.

Figure 4 is an overlay of magnetoresistance as a function of field at various temperatures for field parallel to the conducting plane. Unlike for field perpendicular to the plane, the magnetoresistance displays several different features: (1) the

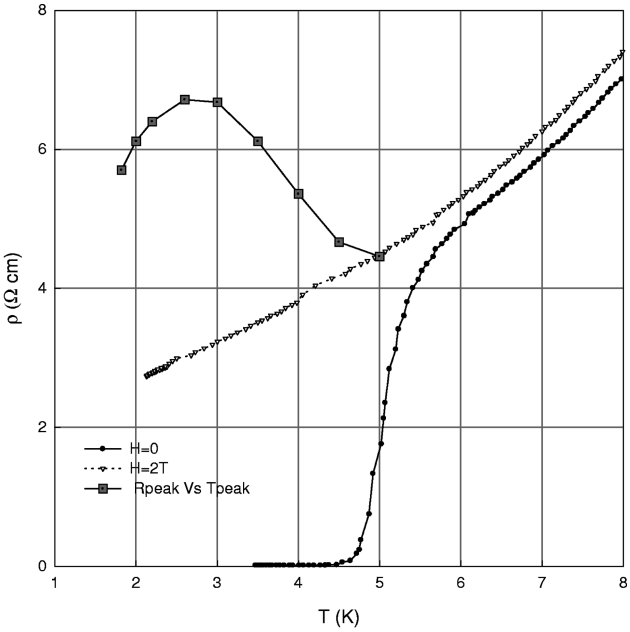

FIG. 3. Overlay of the peak resistivity, resistivity at 2 T, and zero-field resistivity versus temperature.

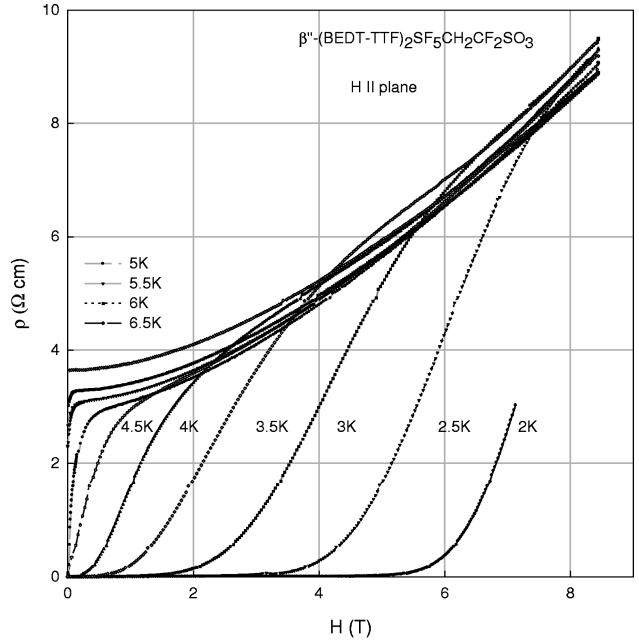

FIG. 4. Interlayer resistivity as a function of field for $H\|$plane at low temperatures. The temperature increment is 0.5 K.

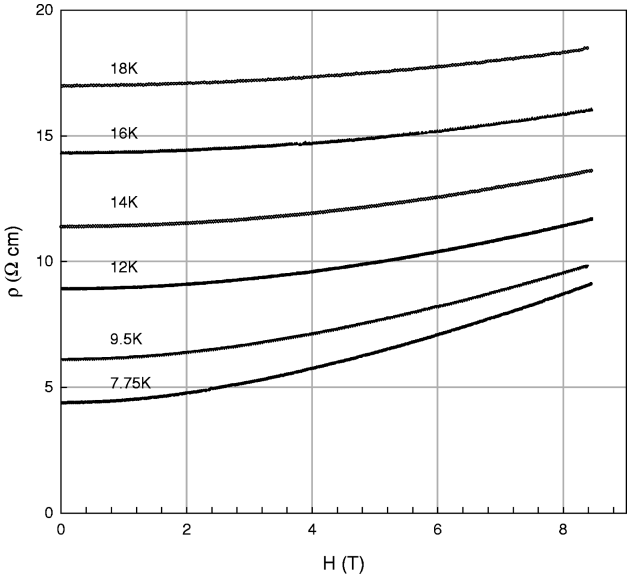

FIG. 5. Interlayer resistivity as a function of field for $H\|$plane at high temperatures.

resistive onset field is much larger for $H\|$plane, for example, $H_{onset}\sim 5$ T at 2 K, compared to $H_{onset}\sim 0.15$ T for $H\perp$ plane; (2) the rise of resistivity for $H>H_{onset}$ is much slower in the $H\|$plane case; (3) there is no peak in resistivity as a function of field. Instead, $\rho(H)$ increases monotonically with increasing field; (4) the magnetoresistance at large fields for $H\|$plane is considerably larger than the peak resistivity value for $H\perp$ plane, similar to the κ-phase salts.[17,18] It should be noted that since the measurements for field parallel and perpendicular to the conducting plane were done in two separate runs, a larger cooling rate for the $H\perp$ plane has resulted in a larger normal-state resistivity and slightly reduced T_c. This is analogous to the cooling rate dependence observed in the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br.[21] If we normalize the zero-field resistivity above T_c to that of $H\|$plane by a scaling factor $[\rho_\|(10\text{ K})/\rho_\perp(10\text{ K})]\cong 0.68$, the normalized peak resistivity at 3 K is $\rho_\perp^{norm}\sim 6.5\times 0.68=4.4\,\Omega$ cm compared with $\rho_\|(8\text{ T})\sim 9\,\Omega$ cm.

At $T>T_c$, magnetoresistance increases with increasing field. Shown in Fig. 5 is an overlay of $\rho(H)$ at various temperatures $T=7.75$, 9.5, 12, 14, 16, and 18 K. With increasing temperature, the field dependence of magnetoresistance is increasingly smaller, as is clear from the figure. The data can be well fitted to $\rho(H)=\rho_o+\Delta\rho^{(2)}H^2+\Delta\rho^{(4)}H^4$, with $\Delta\rho^{(2)}$ and $\Delta\rho^{(4)}$ being the coefficients for H^2 and H^4 terms, respectively.

The temperature and field dependence of the magnetoresistance can be summarized by plotting the temperature dependence of the fitted $\Delta\rho^{(2)}$ and $\Delta\rho^{(4)}$ values scaled to ρ_o, as shown in Fig. 6 in a semilog scale. Clearly, $(\Delta\rho^{(2)}/\rho_o)$ increases exponentially with decreasing temperature. The solid line is a fit to $(\Delta\rho^{(2)}/\rho_o)=0.16\times 10^{-T/8}$. $\Delta\rho^{(4)}$ is always negative for the temperature range investigated, $-(\Delta\rho^{(4)}/\rho_o)$ is plotted against temperature in the inset. Again, an exponential temperature dependence is seen with $(\Delta\rho^{(4)}/\rho_o)=-0.06\times 10^{-T/5.4}$. For temperature above 20 K, $\Delta\rho^{(4)}$ term has a negligible contribution to the magnetoresistance.

An alternative way to look at the field dependence of the magnetoresistance is to measure the temperature dependence of resistivity at a fixed field. Shown in Fig. 7 is an overlay of the $\rho(T)$ at various applied fields $H=0$, 1, 2, 4, 6, and 8 T for $H\|$plane. For $T>6$ K, $\rho(T,H)$ increases monotonically with T. For $T<T_c$, $\rho(T,H)$ has a maximum at a field dependent peak temperature. The results are equivalent to the isothermal field dependence in Fig. 4, except here the peak is better defined due to small temperature increments. As mentioned earlier, the zero-field resistivity for $H\|$plane is smaller than that for $H\perp$ plane and the T_c is about 0.2 K higher. The cooling rate dependence of the resistivity and the reduced T_c with increasing ρ are analogous to that of

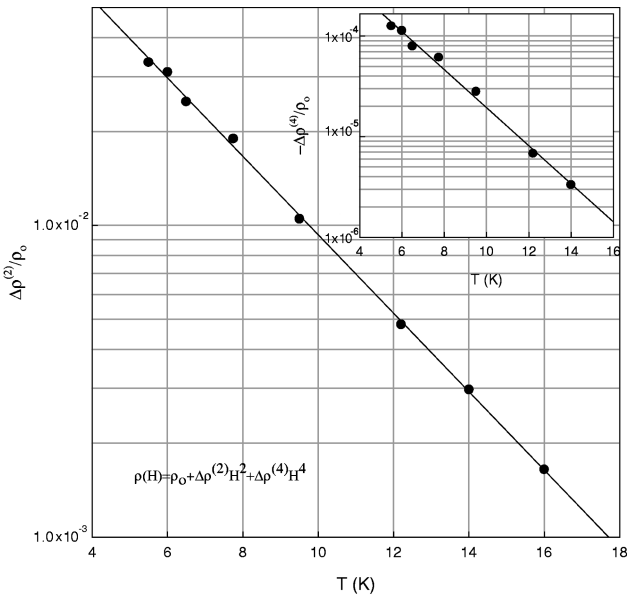

FIG. 6. The extrapolated field and temperature coefficients versus temperature in a semilog scale.

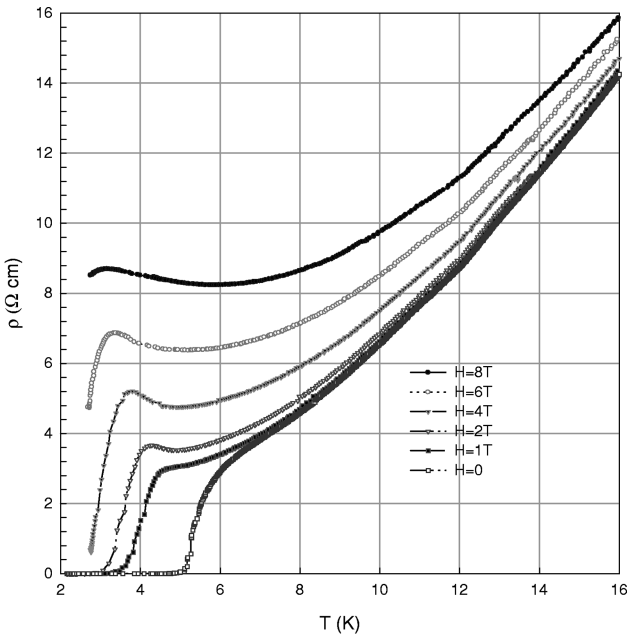

FIG. 7. Overlay of interlayer resistivity as a function of temperature for various applied field for $H\|$plane.

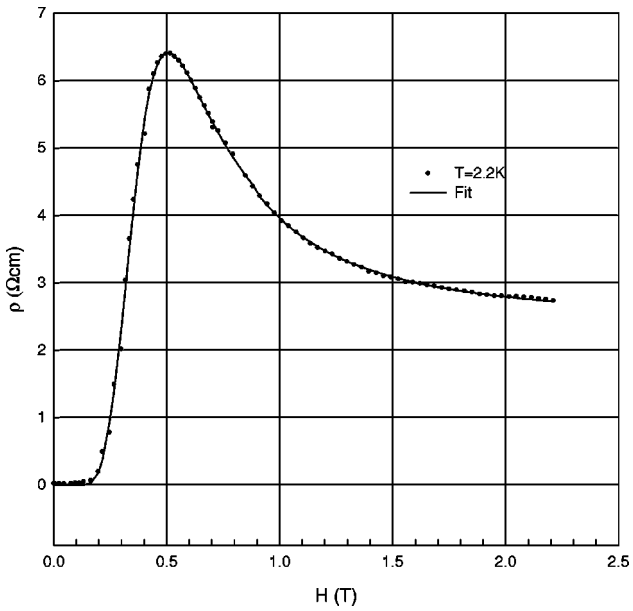

FIG. 8. Interlayer resistivity as a function of field at 2.2 K. The line is a fit to the data.

κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br salt.[21]

The field and temperature dependence of the interlayer resistivity in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ is very similar to that of κ-(BEDT-TTF)$_2X$ with X = Cu[N(CN)$_2$]Br and Cu(NCS)$_2$, even though the superconducting transition temperature in the κ phase is much larger at 11 and 10 K, respectively. The peak effect or large negative magnetoresistance in the $H\|I\perp$ plane is still controversial. One possibility is that the negative magnetoresistance arises from the presence of magnetic impurities in the samples.[11] With increasing field, the magnetic scattering is suppressed, thus leading to a decrease in magneto-resistivity. However, the presence of magnetic impurities in the all-organic β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ is very unlikely, because it has no metal atoms, such as the Cu(II) in the κ-phase structure. The absence of negative magnetoresistance of the in-plane resistivity for high quality κ-phase samples, where peak effect in the interlayer resistance persists, suggests strongly that the magnetoresistance peak may be intrinsic to the layered systems.[13] This is also supported by a recent study of magnetoresistance peak as a function of inhomogeneities in the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br salts.[14] For samples with large superconducting transition widths, the peak effect disappears. With increasingly smaller transition width the peak becomes more pronounced. Negative magnetoresistance can also arise from weak localization and electron-electron interactions as observed in many metallic systems.[22] Disorders can lead to negative magnetoresistance because the magnetic field disrupts the coherent backscattering and suppresses the localization. Similarly, the magnetic field will decrease the attractive electron-electron interaction and lead to a smaller resistance for charge transport. However, the magnitude of the peak or $(\Delta\rho/\rho)\sim 1$ is too large to be considered for this model. A magnetoresistance peak is also possible if one assumes a large vortex-lattice interaction.[10] In this model, vortex-lattice interaction leads to local disorders in the electronic potentials, thus giving rise to an extra scattering. With increasing field, the vortex cores start to overlap. Eventually, the large field will reduce the lattice distortions and resume the normal-state electric conductivity. Although the model is plausible, there has been no report of structural evidence for the lattice distortions.

Negative magnetoresistance has been discussed recently in terms of stacked Josephson-junction model including a field dependent quasiparticle tunneling.[12–14] In this model, the resistive transition at small fields can be described by a resistively shunted Josephson junction with $R(H) = R_n[I_o(\hbar I_c/2ekT)]^{-2}$, where R_n is the normal-state resistance, $\hbar$ is the Planck's constant, I_c is the critical current, e is the charge of an electron, and I_o is the modified Bessel function. For $H\perp$ plane, the junction is effectively determined by the distance between the neighboring vortices. In general, the quasiparticle conductance Y_{ss} is thermally activated $Y_{ss} \sim \exp[-\Delta(T,H)/kT]$, and the pair conductance is $Y_p \sim [I_o(\hbar I_c/2ekT)]^2 - 1$ The total conductance is $Y = Y_{ss} + Y_p = 1/\rho$. With increasing field, the pair contribution decreases while the quasiparticle contribution increases. The competition among the two terms naturally leads to a peak in the conductance or magnetoresistance.

To analyze it quantitatively, charge transport is considered to be along an effective Josephson junction of area $a^2 \approx (\Phi_o H + H_o)$ between the densely packed vortices,[4] H_o being a fitting parameter. The total junction conductance is given by $Y = Y_1(\{I_o[e_J\Phi_o/(H+H_o)2kT]\}^2 - 1) + Y_2(H)\exp[-\Delta(T,H)/kT]$, here $e_J = (\hbar I_c/2ea^2)$ defines an intrinsic Josephson coupling energy, $Y_1 = 1/R_n$. If the field dependence of the e_J and $\Delta(T,H)$ is considered to be proportional to $(1-[H/H_{c2}(T)]^2)$ and assume a constant Y_2, we are unable to get a reasonable fit to the data. If we assume $1/Y_2(H) = \rho_0[1+(Ho/H)^n]$, a nearly perfect fit near the peak can be obtained, as shown in Fig. 8. The fit gives $1/Y_1 = 6.9 \pm 0.1\ \Omega$ cm, $(e_{oJ}\Phi_o/2kT) = 0.46 \pm 0.02T$, $H_o = -0.03 \pm 0.005T$, $1/Y_2(H) = 2.34 + (1.53/H^2)\ \Omega$ cm, $(\Delta_o/kT) = 0 \pm 0.01$, $H_{c2} = 0.9T$ for ρ(H) at $T = 2.2$ K.

Clearly, the exponential term in the quasiparticle contribution is negligible. Fits without the exponential term give the same parameters. Similar results are obtained for other temperatures. This is consistent with the fact that the negative magnetoresistance extends well above H_{c2}. Unless a very different field dependence of the gap energy is considered, such as $\Delta(T,H)\sim 1/H^{\alpha}$, a simple model considered above fails to support the picture where the negative magnetoresistance comes from the enhanced quasiparticle tunneling. One possibility is to include the fluctuation effect for $H>H_{c2}$ in the treatment of quasiparticle contribution. It should be noted that in a similar approach to fit the peak effect in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, the model failed to fit the high-field data.[12]

For field applied parallel to the plane, the field will be mostly confined in between the superconducting layers. The critical field in this direction is much larger than that when the field is perpendicular to the plane. The larger resistive onset field is a direct consequence of the large anisotropy in this material. The large positive magnetoresistance at high field and high temperatures may be associated with the open sheets and closed pockets in the Fermi surface. For most metals, the magnetoresistance is negligible with a typical $(\Delta\rho/\rho)$ in the order of $(\omega\tau)^2=[(eH/m)\tau]^2$. For example, in copper $\omega\tau\sim 5\times 10^{-3}$ for a field of 1 T. A similar estimate for the title compound would yield $\omega\tau=\sqrt{(\Delta\rho^{(2)}/\rho_o)}\sim 0.2$ at 1 T. This is almost two orders of magnitude larger than in conventional metals. However, the interlayer resistivity near the transition is about 1 Ω cm, about six orders of magnitude larger than in copper. The large $(\Delta\rho/\rho)$ demonstrates the gross inadequacy of a single band picture. In the presence of two bands, as in the case of most organic conductors, a large positive magnetoresistance is foreseeable.[23,24] Consider n_1 and n_2 as the charge densities for the two bands and μ_1 and μ_2 as the carrier mobilities, respectively. The zero-field resistivity is $\rho_o=(n_1\mu_1+n_2\mu_2)^{-1}$. At low fields, the transverse magnetoresistance is $(\Delta\rho/\rho_o)=[n_1n_2\mu_1\mu_2(\mu_1-\mu_2)^2H^2/(n_1\mu_1+n_2\mu_2)^2]$; at higher fields a negative H^4 term should be included in the two band model. It should be noted that the above expression for the transverse magnetoresistance applies only to the isotropic two band system. The anisotropic nature of the present compound, added with uncertainties in the charge carrier densities and mobilities, makes it very complicated task to identify the contribution from each terms. Nevertheless, if we assume $\mu_1\gg\mu_2$, then $(\Delta\rho/\rho_o)\approx(n_2\mu_1\mu_2/n_1)H^2$. The exponential temperature dependence in the $(\Delta\rho^{(2)}/\rho_o)=0.16\times 10^{-T/8}$ for $H=1$ T suggests that $\mu_1\mu_2\sim 10^{-T/8}$. A similar exponential temperature dependence might be expected in $(\Delta\rho^{(4)}/\rho_o)\sim 10^{-T/5.4}$, since it involves higher orders of $\mu_1\mu_2$. While a conventional Fermi-liquid system gives a power-law temperature dependence in $(\Delta\rho^{(2)}/\rho_o)$, the experimental results may suggest the exponential temperature dependence is due either to complications of in-plane anisotropy or non-Fermi-liquid behavior.

IV. CONCLUSIONS

In summary, we have reported a detailed interlayer transport measurement in the all-organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. For field perpendicular to the layers, a peak in magnetoresistance is observed as a function of field. The peak field increases with decreasing temperature with $H_{peak}(T)=H_o(1-T/T_c)^2$. For $H>H_{peak}$, a large negative magnetoresistance is observed. Further increase in field results in a small positive magnetoresistance. The interlayer magnetoresistance peak effect is very similar to that of the κ-phase organic superconductors. This demonstrates that the peak effect is intrinsic to the layered organic superconductors. Quantitative analysis in terms of stacked Josephson-junction model plus a thermally activated quasiparticle tunneling is unable to fit the negative magnetoresistance, assuming a simple field dependence of the gap energy $\Delta(T,H)=\Delta_o\{1-[H/H_{c2}(T)]^2\}$. The origin of the peak effect in the interlayer transport remains unclear. For field parallel to layers, large positive magnetoresistance is observed for all temperatures with large resistive onset field for $T<T_c$. Although the data are generally consistent with two band picture for the organic conductors, the exponential temperature dependence of $(\Delta\rho^{(2)}/\rho_o)$ calls for more systematic studies of the anisotropic transport in the conducting plane.

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