Interlayer magnetoresistance in the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br near the superconducting transition

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In this paper, we report transport measurements of interlayer magnetoresistance with field parallel and perpendicular to the current direction in the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. For $H \| J$, the isothermal magnetoresistance $R(H)$ displays a peak effect as a function of field. While the magnetoresistance at small field can be fitted to stacked Josephson junction model, the negative magnetoresistance is not consistent with quasiparticle tunneling model with a simple mean field gap. The origin for the peak effect remains unresolved. For $H \perp J$, $R(H)$ increases monotonically with increasing field. Large magnetoresistance for $H \perp J$ is consistent with the layered structure of the organic compounds. [S0163-1829(99)03025-8]

I. INTRODUCTION

Charge transport in the direction perpendicular to the superconducting layers of the cuprates has been of recent interest.[1–3] The temperature dependence of c-axis resistivity depends strongly on the oxygen concentration, changing from a metallic temperature dependence in the optimally doped samples to a semiconducting behavior in the underdoped.[4,5] Magnetoresistance in this direction shows a pronounced peak as a function of temperature and field before it drops to zero.[6–8] This has been discussed extensively in the very anisotropic oxide $Bi_2Sr_2CaCu_2O_8$ superconductors[9–15] and in the oxygen deficient $YBa_2Cu_3O_{7-\delta}$ materials.[12] The results can be qualitatively interpreted in the framework of stacked Josephson junctions between the superconducting layers. The peak in the magnetoresistance at fixed field is attributed to the competition between the pair and the quasiparticle tunneling. The organic superconductors κ-(BEDT-TTF)$_2X$ [bis(ethylenedithio)tetrathiafulvalene, abbreviated as ET] with X being Cu[N(CN)$_2$]Br$^-$ and Cu(SCN)$_2^-$, with the conducting cation layer (ET) separated by the insulating anion layers (X) have shown similar anisotropic transport properties.[16,17] Furthermore, interlayer transport studies show an interesting magnetoresistance peak effect as a function of field and temperature in the mixed state.[18–22] However, there are several important differences among the two systems. One is the temperature dependence of the normal state resistance. While the cuprates typically have semiconducting temperature dependence in the c axis for the underdoped, the interlayer resistance in the organics has metallic temperature dependence near the superconducting transition. Another major difference is the large negative magnetoresistance observed in the organic compounds, which gives rise to a pronounced peak resistance as a function of field. While there are several models proposed for the peak effect, such as the magnetic impurity scattering[18] and vortex-lattice interaction.[19] A similar mechanism as in the cuprates, involving pair and quasiparticle tunneling has been proposed by several groups.[20–22]

To understand the interlayer charge transport, we have performed careful measurement on newly discovered rod samples of κ-(ET)$_2$Cu[N(CN)$_2$]Br with field up to 18 T. Unlike the conventional platelet geometry of the organic superconductor, where the shortest dimension (b axis) is perpendicular to the conducting planes (ac plane). The sample length in b-axis direction in the rod sample is the largest. The availability of the rod sample makes it possible for an ideal four probe measurement of the interlayer resistance.

In this paper, we report interlayer magnetoresistance measurement with field parallel and perpendicular to the b axis. For field parallel to b axis, similar results as reported on plate samples are observed. Magnetoresistance $R(H)$, with field H parallel to the current, displays a peak as a function of field and temperature. The low field magnetoresistance can be well fitted to a Josephson junction model. However, the negative magnetoresistance cannot be fitted to a simple thermally activated quasiparticle tunneling. This raises a serious question about the dominant role of quasiparticle in the phenomena of negative magnetoresistance. For field perpendicular to the current, i.e., H being in the ac plane, several different features in $R(H)$ are observed. First, the characteristic field in this orientation is much larger that the parallel direction. Second, no obvious peak in $R(H)$ is observed and the characteristic resistance increases monotonically with decreasing T. Third, $R(H)$ increases quadratically without saturation at large H. The findings are consistent with the layered nature of the organic superconductor being studied.

II. EXPERIMENT

Single crystals of the κ-(ET)$_2$Cu[N(CN)$_2$]Br superconductor were synthesized by the electrocrystallization technique described elsewhere.[23] Several crystals were used in these measurements with average dimensions of $1-2\times0.5\times0.5$ mm with the superconducting transition temperature near 12 K. Interlayer resistivity near T_c is around 1Ω cm. The largest length is along the least conducting direction. High field (up to 18 T) measurements were performed at the National High Magnetic Field Laboratory. Similar measure-

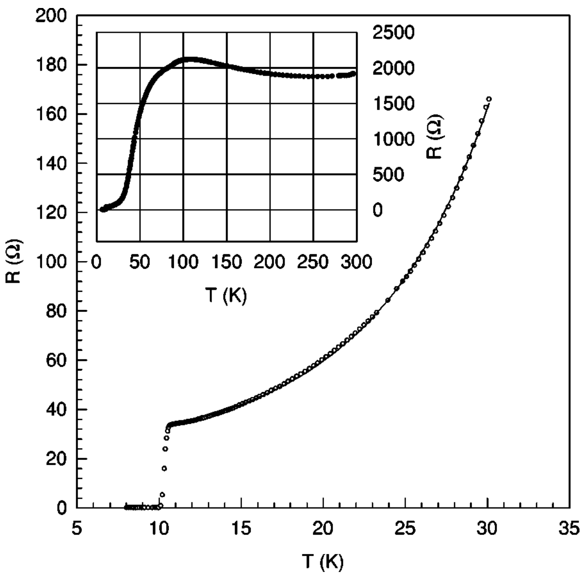

FIG. 1. Interlayer resistance as a function of temperature near T_c for $H \| J$ and the solid line is a fit. The inset is the $R(T)$ for the whole temperature range.

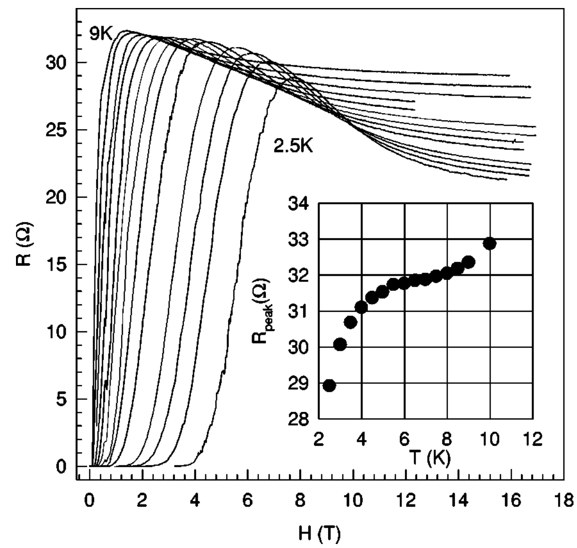

FIG. 2. Magnetoresistance as a function of field for $H \| J$ at different temperatures ($T = 2.5 - 9$ K with 0.5 K increment). The inset is the R_{peak} as a function of T.

ments with a smaller magnet (up to 9 T) yielded the same results. 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 31 Hz. The samples were cooled slowly to below the superconducting transition temperature with the field parallel and perpendicular to the crystallographic b axis. For measurements at different orientations, the sample was warmed up to room temperature and remounted. A misalignment of up to 5° degrees was possible due to the small sample size.

III. RESULTS

A. $H \| J \perp$ plane

Shown in Fig. 1 is a typical temperature dependence of interlayer resistance near the transition temperature. The resistance decreases rapidly at high temperatures and tends to saturate near the transition. The solid curve through the data points is a fit to $R(T) = R_0 \exp[(T/T_0)^2]$ with $T_0 = 22.5$ K. It should be pointed out that the exponential T^2 dependence can only be fitted to the data in the temperature range below 30 K. At higher temperatures, especially near the inflection point near 40 K, the curve starts to deviate from the data. The data can also be fitted to an power-law temperature dependence $R(T) = a + bT^n$, with n being dependent on the temperature range. For example, for the same temperature range, $n = 3.88$ gives the best fit. Nevertheless, the overall quality with power-law fitting is not as good as the exponential T^2 fit.

The inset shows the overall temperature dependence of the resistance from room temperature down. Similar to work reported earlier,[23] the resistance decreases initially with temperature to about 250 K. Below 250 K, a semiconducting temperature dependence is observed with the resistance increases with decreasing T. A broad peak in R is usually observed near 100 K. Depending on the cooling rate, a resistance anomaly at 80 K has been reported.[24] For temperature below 80 K, R decreases rapidly with T with a change of curvature near 40 K.

Figure 2 is an overlay of magnetoresistance as a function of field for the $H \| J$ geometry for 2.5 K $\leq T \leq$ 9 K. The curves from right to left correspond to temperatures from 2.5 to 9 K with an increment of 0.5 K. At each fixed temperature, the interlayer resistance becomes finite at some onset field and increases rapidly with increasing H until it reaches a peak R_{peak} at H_{peak}. For $H > H_{peak}$, the magnetoresistance becomes negative and saturates at further higher fields. With increasing temperature, the dissipative onset field decreases with a corresponding decrease in H_{peak}. Magnetoresistance at different temperatures crosses each other at two fields near the peak field. However, the magnetoresistance at very high fields (above 10 T) has a monotonic temperature dependence with $R(H)$ increasing with increasing T.

The inset in Fig. 2 is a plot of the peak resistance as a function of temperature. A shoulderlike feature in $R_{peak}(T)$ is clearly observed near 6 K. Below 5 K, R_{peak} decreases rapidly with decreasing T.

An alternative way to look at the field and temperature dependence of the interlayer magnetoresistance is to plot the magnetoresistance as a function of temperature at fixed field, as shown in Fig. 3. At small fields, $R(T)$ displays a similar peak effect. For example, at $H = 4$ T, a peak in $R(T)$ is clearly seen at around 5.5 K. With increasing H, the peak temperature is suppressed. At 10 T, no observable peak is in sight within the temperature range.

The temperature dependence of the peak field can be summarized in Fig. 4. Here peak fields measured on two different samples are overlaid with the square symbol corresponding to the data reported here. The two sets of data show a clear overlap. The solid line is a fit to the expression $H_{peak} = H_0(1\text{-}T/T_c)^m$ with $H_0 = 10.4 \pm 0.3$ T, $T_c = 11.5 \pm 0.5$ K, and $m = 1.5 \pm 0.1$. The fitted T_c agrees with the zero field data in Fig. 3.

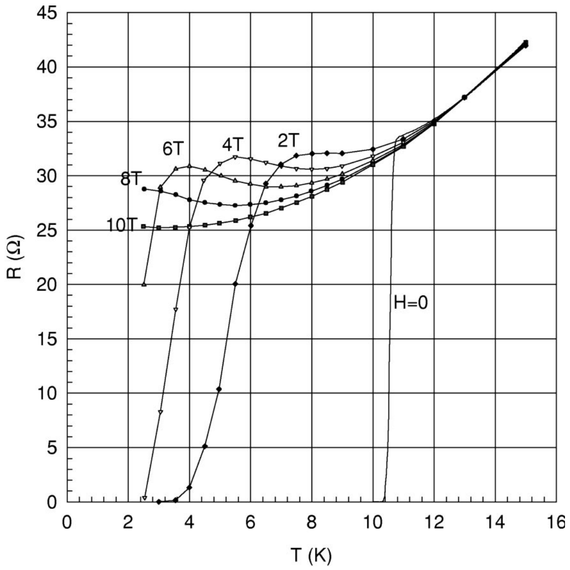

FIG. 3. Magnetoresistance as a function of temperature for $H \| J$ at different fields $H = 0,2,4,6,8,10$ T.

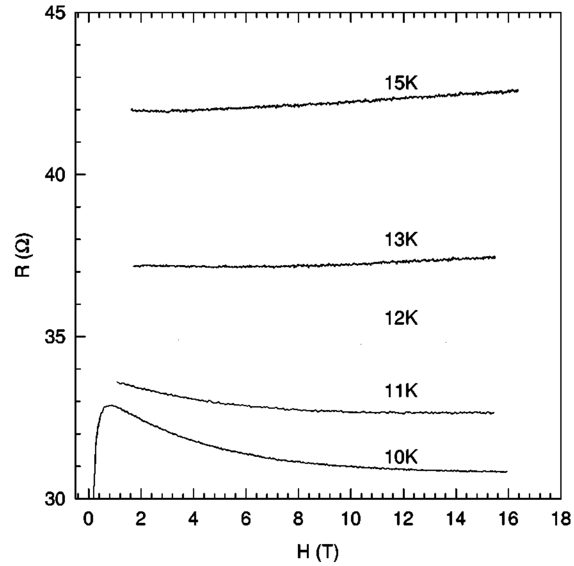

FIG. 5. Magnetoresistance as a function of field at high temperatures $T = 10,11,12,13,15$ K.

In the normal state ($T \geqslant 11$ K), the field dependence of magnetoresistance is drastically reduced, as shown in Fig. 5. For comparison, $R(H)$ at $T = 10$ K, where peak is observed, is included as well. With increasing temperatures, a crossover from negative magnetoresistance to positive magnetoresistance is observed. At $T = 11$ K, dR/dH is negative for field less than 16 T. At $T = 12$ K, dR/dH changes from negative to positive at around 9 T. At $T = 13$ K, dR/dH is almost $\geqslant 0$ for all fields, as is the case for $T = 15$ K with a $dR/dH \simeq 0.05$ Ω/T at high H.

B. $H \|$ plane $\perp J$

Similar measurements have been performed with the field applied parallel to the planes, as shown in Fig. 6. The sixteen curves correspond to temperature at 2.5 K to 10 K at an increment of 0.5 K, from right to left. In contrast to the results presented in Fig. 1, no peak in $R(H)$ is observed for all temperatures. At a fixed temperature, the magnetoresistance rises sharply for field above an onset field. $R(H)$ reaches a plateau for a range of intermediate fields and it rises again at higher fields. The characteristic fields increases with decreasing temperatures. The resistive onset field is much greater than that for $H\perp$ plane. For example, at $T = 2.5$ K, $H_{\rm onset}$ is about 13 T for $H \perp J$, compared with 4 T for $H \| J$.

The temperature dependence of the interlayer magnetoresistance at fixed field can be obtained from the $R(H)$ data. Shown in Fig. 7 is an overlay of $R(T)$ at different field $H = 0,1,2,3,6,8,10,12,15$ T, from right to left. The peak in $R(T)$ is a nature consequence of the reduced magnetoresistance at lower temperature. It should be noted that the zero field transition temperature is noticeably reduced from the previous measurement geometry. The difference in T_c is due

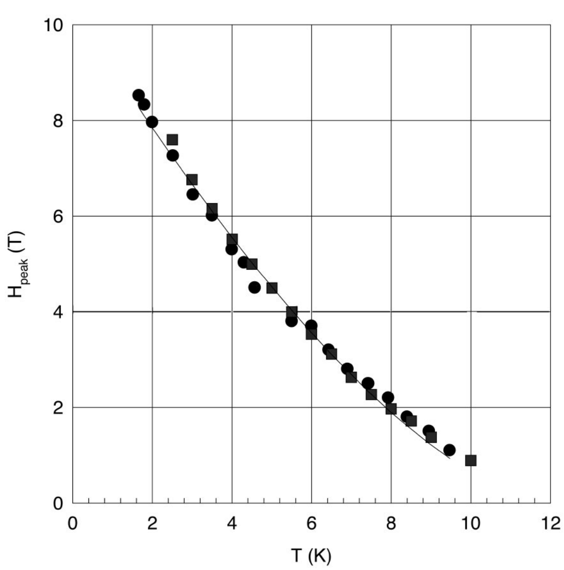

FIG. 4. Peak field as a function of temperature for two samples. The solid line is a fit to the data.

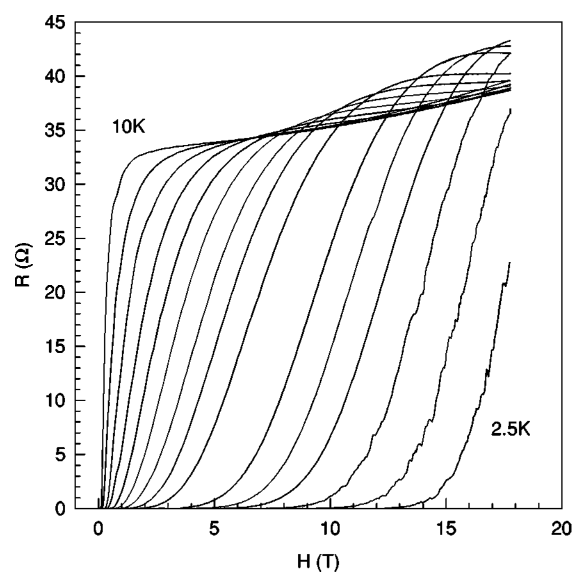

FIG. 6. Magnetoresistance as a function of field at different temperatures for $H \perp J (T = 2.5 - 9$ K with 0.5 K increment).

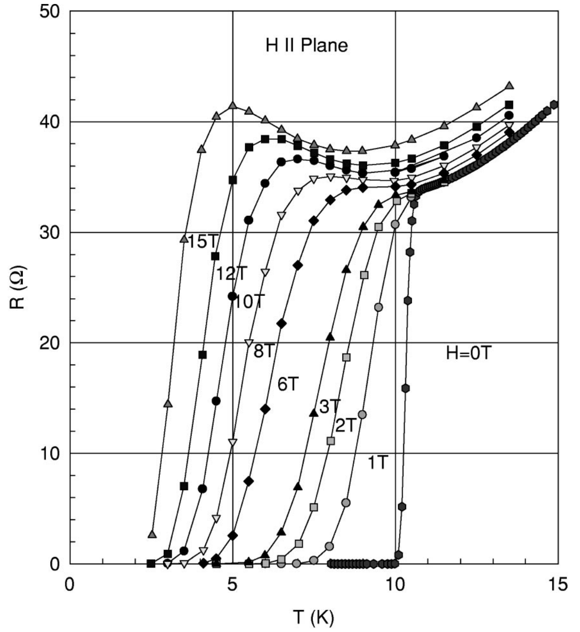

FIG. 7. Magnetoresistance as a function of field for $H \perp J$ at high temperatures, $T = 10, 10.5, 11.5, 12.5$, and 13.5 K. The inset is a comparison for $H \perp J$ and $H \| J$ at 12.5 K and 13 K, respectively.

to a different cooling rate for the two measurements. A detailed study of the cooling rate dependence of the superconducting transition temperature has been reported earlier. The reduced T_c is consistent with the faster cooling after the sample is realigned for $H\|$ plane.

At temperatures above the transition, a large positive dR/dH is found, as shown in Fig. 8. With increasing temperature, $R(H)$ curves shift upward. For each fixed temperature, the field dependence can be well fitted with an expression $R(H) = a + bH^2 + cH^4$. The coefficients b and c are nearly constants with $b = (2.5 \pm 0.1) \times 10^{-2}$ and $c = (1.0 \pm 0.2) \times 10^{-5}$, as the curves are nearly parallel. The inset is a comparative plot of the magnetoresistance for both orientations at 12.5 K and 13 K. The increase in resistance in the same field range is about 10 times or more larger for $H \perp J$ than that for $H \| J$.

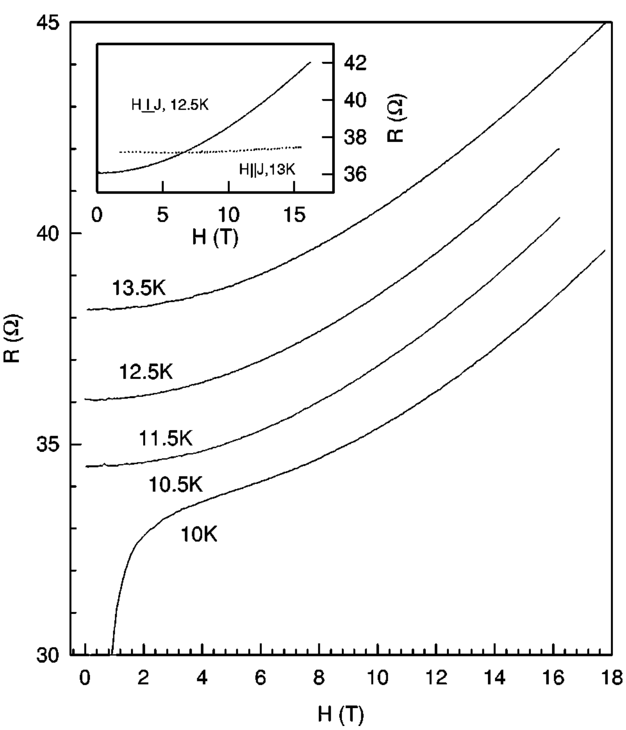

FIG. 8. Magnetoresistance as a function of temperature for $H \perp J$ at different fields $H = 0, 1, 2, 3, 6, 8, 10, 12, 15$ T.

IV. ANALYSIS

The field dependence of magnetoresistance in the geometry of $H \| J \| c$ in the mixed state has been studied extensively in the highly anisotropic cuprates, namely, $Bi_2Sr_2CaCu_2O_8$ and the oxygen deficient $YBa_2Cu_3O_{7-\delta}$. Experimentally, the majority of the published work shows magnetoresistance as a function of temperature at fixed fields. In the case of $Bi_2Sr_2CaCu_2O_8$, the peak effect in the $R(T)$ has been universally reported. However, differences among published results in $R(H)$ exist upon careful examination. For example, work reported on some single crystal samples shows no peak in $R(H)$,[10–13] while a peak in $R(H)$ can be easily constructed for some other samples.[6–8,14] In the case of organic superconductors studied here, x-ray diffraction at ambient temperature shows no evidence of second phases. A recent study of the peak effect as a function of resistive transition width shows that the peak effect disappears gradually as the transition width increases, demonstrating clearly that the peak in $R(H,T)$ is intrinsic only to the high quality samples in the organic superconductors.[22] Although the layered structures are very similar, the interlayer transport may be substantially different. For example, in the cuprate, the peak resistance as well as the normal state resistance, increases monotonically with decreasing temperature near T_c. On the contrary, the interlayer resistance of the organic superconductors studied here decreases with decreasing temperature.

The dissipation mechanism in the interlayer direction in the mixed state remains controversial. Two approaches are known to give rise to a peak in the interlayer resistivity as a function of temperatures. One of them models the resistivity peak as a result of fluctuations above the mean field transition temperature.[25,26] Of the four possible fluctuation contributions to the interlayer resistivity, fluctuations in the density of state (DOS) and the regular Maki-Thompson term contribute to an increasing resistivity with decreasing temperatures. By choosing suitable parameters, the model can fit reasonably the temperature dependence of the resistivity before the peak for the cuprates. However, the model does not include critical fluctuations nor contributions from the vortex state (for $T < T_{peak}$) and thus the field dependence of the peak temperature $T_{peak}(H)$. A more widely adopted approach, discussed in the following, emphasizes the nature of Josephson coupling between the superconducting layers, especially at low magnetic fields. The peak in the resistivity comes when the Josephson coupling dominates the interlayer transport. The model can describe semiquantitatively the field and temperature dependence of the interlayer transport at $H < H_{peak}$.

In the case of $H \| J \| c$, dissipation mechanism was first proposed by Briceño *et al.*[9] In this model, current moving parallel to the c axis is taken to pass through a narrow superconducting channel of area $A \approx \Phi_o / H$ between the densely packed vortices. Here Φ_0 is the flux quantum. Dissipation occurs through thermodynamic fluctuations which cause the phase of the superconducting order parameter in the c direction to jump by 2π. Assuming fluctuations in each channel are independent, the dissipation in the c direction can be modeled by a long, narrow Josephson junction at finite T.[27] The resistance of the weak link is given approximately by $R = R_n[I_0(\hbar I_c/2ekT)]^{-2}$, where R_n is the normal state resistance, $\hbar$ is the Planck constant, I_c is the critical current, e is the charge of an electron, and I_0 is the modified Bessel function. Since the normal state resistance is activated in this direction, a peak is expected in the junction resistance at $T < T_c$. A similar approach which models the c axis conduction as a stack of Josephson tunnel junctions has been proposed by Gray *et al.*[10] For an intermediate Josephson coupling, the junction conductance is the sum of the quasiparticle conductance Y_{ss} and pair conductance Y_p, i.e., $Y = Y_{ss} + Y_p$. Since the quasiparticle conductance Y_{ss} is thermally activated $Y_{ss} \sim \exp[-\Delta(T,H)/kT]$, and the pair conductance $Y_p \sim [I_0(\hbar I_c/2ekT)]^2 - 1$, a distinct peak in $R(T)$ arises naturally. While the field dependence of the crtical current I_c is somewhat controverital,[11] a recent study on thin mesa of a $Bi_2Sr_2CaCu_2O_8$ found $I_c \propto 1/H$.[15]

A similar model has been proposed to explain the magnetoresistance peak in the organic superconductors.[20–22] To analyze the data quantitatively, consider the charge transport through a Josephson junction of area $a^2 \approx \Phi_0/H + H_0$ between the densely packed vortices, H_0 being a fitting parameter to take into account of effects of pinning and lattice rigidity.[12] The junction resistance due to thermal fluctuations of the phases is given by $R(H) = R_{n'}[I_0 E_J/2kT]^{-2}$, where $E_J = \hbar I_c/e = [\pi\hbar\Delta(T)/2e^2 R_{n'}]\tanh[\Delta(T)/2kT]$ is the Josephson coupling energy, $R_{n'}$ is the junction resistance in the mixed state. Because I_c is proportional to the junction area, E_J is also. It is natural to define an intrinsic Josephson coupling energy $e_J = E_J/a^2$, such that $R(H) = R_{n'}\{I_0[e_J\Phi_0/(H + H_0)2kT]\}^{-2}$. If $E_J \gg kT$, the junction resistance can be reduced to[12] $R(H) = R_{n'}\exp[-e_J\Phi_0/(H+H_0)kT]$. However, the $E_J \gg kT$ is limited only to very small field. To fit the $R(H)$ over the whole field range, a tabulated Bessel function I_0 has to be used. The total junction conductance $Y = Y_1(\{I_0[e_J\Phi_0/(H+H_0)2kT]\}^2 - 1) + Y_2(H)\exp[-\Delta(T,H)/kT]$.

To fit the data, we have assumed a simple field dependence for coupling energy $e_J = e_0 J[1-(H/H_{c2})^2]$ and the gap energy $\Delta(T,H) = \Delta_0[1-(H/H_{c2})^2]$.[28] If we assume a constant Y_2 for the quasiparticle contribution, the data cannot be fit at all. Assuming $1/Y_2(H) = a + b/H$, a fit can be obtained as shown in Fig. 9 for $R(H)$ at $T = 5$ K. The fitting parameters are $1/Y_1 = 82 \pm 1\,\Omega$, $e_0 J\Phi_0/2kT = 5.8 \pm 0.2$ T, $H_0 = 0.3 \pm 0.1$ T, $1/Y_2(H) = 20 + (63/H)\Omega$, $\Delta_0/kT = 0.06 \pm 0.02$, $H_{c2} = 6.4$ T. The overall fit is rather satisfactory, however, the gap energy thus fitted is physically negligible. If we neglect the thermal activation term with $1/Y_2(H) = R_0[1+(H_1/H)^\alpha]$, an equally well fit can be achieved with $R_0 = 20 \pm 1\,\Omega$, $H_1 = 3.2 \pm 0.2T$, $\alpha = 1.3 \pm 0.1$. The inset shows the direct fit to the resistively shunted Josephson Junction model at small fields $R(H) = R_{n'}\{I_0[e_J\Phi_0/(H + H_0)2kT]\}^{-2}$. The fit gives $R_{n'} = 94\ \Omega$, $e_J\Phi_0/2kT = 5.2$ T, and $H_0 = 0.2$ T. The comparable parameters for

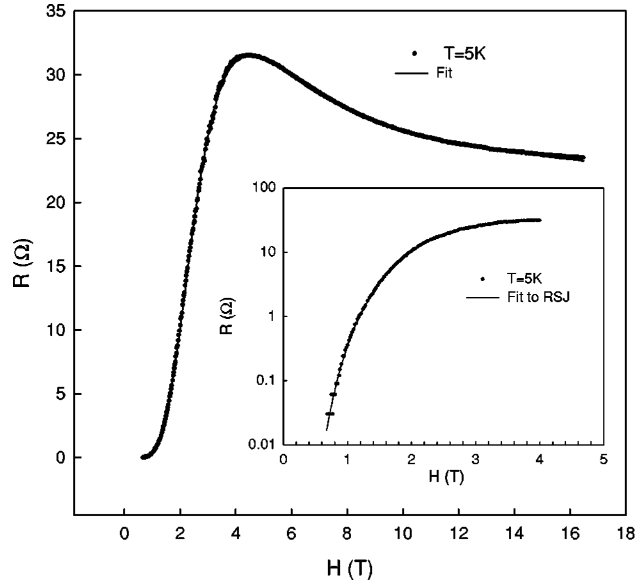

FIG. 9. Magnetoresistance as a function of field for $H \parallel J$ at 5 K. The solid line is a fit to the model. The inset is an expanded view at small fields. The line is a fit to the RSJ model.

the Josephson coupling energy obtained from small field fit and over the whole field range gives confidence in the uniqueness of the fitting parameters obtained.

Similar fit can be obtained for magnetoresistance at other temperatures. Using $1/Y_2(H) = R_0[1+(H_1/H)^\alpha]$, the exponent α is found to increase with decreasing temperature. For example, at $T = 3$ K, $\alpha \sim 2$ and it decreases to ~ 0.5 at 7 K. Inclusion of the exponential term in the quasiparticle contribution will yield a near zero gap energy. This is contrary to the picture where the dominant role in the magnetoresistance peak is played by the thermally activated quasiparticles. Rather, it suggests that the negative magnetoresistance is not dominated by the suppression of gap by field. Indeed, it would be difficult to see why the negative magnetoresistance extends over to a field ~ 17 T, much higher than $H_{c2} \sim 6$ T at 5 K. This is also consistent with the report that the high field magnetoresistance in the κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ salt cannot be fit.[21] A modification to the simple field dependence of the gap energy, such as $\Delta(T,H) \propto 1/H^\alpha$, is necessary to fit the data. It is not clear how the various fluctuation terms can affect the quasiparticle contribution above T_c or H_{c2}, further analysis including the fluctuation effect is thus highly desirable.[25,26]

The negative magnetoresistance can also arise from other interactions. For example, it has been proposed that the vortex-lattice interaction could introduce lattice distortions. The lattice distortions provide additional scatterings for the charge carriers. With increasing field, vortices will start to overlap, leading to the restoration of the normal scattering. Thermal fluctuations of the vortices can give similar negative magnetoresistance. With increasing field, the thermal fluctuation is suppressed and vortices are more rigid or lattice-like, resulting in the negative magnetoresistance. Nevertheless, one would not expect the negative magnetoresistance to persist well above the upper critical field.

Presence of magnetic impurities can also give rise to the peak effect. It has been suggested that the κ-(ET)$_2$Cu[N(CN)$_2$]Br may contain traces of Cu^{++} ions.[18]

With increasing field, the effect of magnetic moment scattering is reduced. However, the absence of negative magnetoresistance in the normal state suggests against the magnetic impurity model. Furthermore, a recent study in the all-organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ shows a very similar peak effect in the superconducting state.[29] Since there are no metal ions present in the material, it is very unlikely that the negative magnetoresistance is related to the magnetic ions.

Interlayer transport for the magnetic fields applied in the orthogonal directions are clearly different. For field applied parallel to the plane, the field will be mostly confined in between the superconducting layers. Unlike the $H\perp$ plane, the Josephson vortices do not have normal cores similar to the pancake vortices. The dissipations due to phase slip or thermal fluctuation of vortices have negligible contributions for the $H\|$ plane case. This results in much larger critical fields such as the resistive onset field and the upper critical field. For highly anisotropic systems such as the organic superconductors, the upper critical field is extremely sensitive to even a small misalignment. The present setup for the $H\|$ plane measurement is limited by the small sample size and the mechanical alignment without a fine tuning. This could lead to a substantial underestimate of the upper critical field. Nevertheless, the very different field dependence of the magnetoresistance in the two directions suggests strongly that they reflect the characteristics of the two orientations.

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 $\omega\tau\sim5\times10^{-3}$ for a field of 1 T. A similar estimate for the title compound would yield $\omega\tau=\sqrt{\Delta\rho^{(2)}/\rho_0}\sim0.02$ at 1 T. This is one order of magnitude larger than in conventional metals. However, the interlayer resistivity near the transition is about 1Ω cm, about six orders of magnitude larger than in copper. The large $\Delta\rho/\rho$ demonstrates the gross inadequacy of a isotropic, single band approach. There are two possible mechanisms for the large magnetoresistance. One is due to the intrinsic layered structure. If the transport perpendicular to the planes is coherent, the Fermi surface takes the form of a warped cylinder. With field applied in the plane, it is found that a H^2 dependence is expected for both low and high fields with no saturation in magnetoresistance.[30] For the intermediate regime as defined by $1\leqslant\Omega\tau\leqslant\sqrt{\varepsilon_F/t_\perp}$, where $\Omega=eHv_\| c/\hbar, t_\perp$ is the interlayer transfer integral, and ε_F the Fermi energy, a linear field dependence of $\Delta\rho/\rho$ is predicted. The present observation of quadratic field dependence would suggest $\Omega\tau\leqslant1$. An alternative approach is to consider the effects of two bands in the plane and conduction in the interlayer direction is diffusive.[31,32] 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 $\rho_0=(n_1\mu_1+n_2\mu_2)^{-1}$. At low fields, the transverse magnetoresistance is $\Delta\rho/\rho_0=n_1n_2\mu_1\mu_2(\mu_1-\mu_2)^2H^2/(n_1\mu_1+n_2\mu_2)^2$; at higher fields a negative H^4 terms should be included in the two band model. While both models can qualitatively describe the large interlayer magnetoresistance, a careful angular dependence of field in the plane will be necessary to resolve the issue, as the former should be isotropic with field in the plane and the later be very anisotropic. It should be mentioned that a similar large, positive magnetoresistance has been observed in another organic superconductors β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ in this configuration,[29] suggesting it to be a generic feature of the layered systems.

V. CONCLUSIONS

We have carried out careful interlayer magnetoresistance measurements on the organic superconductor. For field parallel to the current direction, the magnetoresistance exhibits a peak as a function of field and temperature. The peak field increases with decreasing temperature. The field dependence of the magnetoresistance is analyzed in terms of the stacked Josephson junction model with both pair and quasiparticle tunneling. However, a simple mean field model gives an unrealistic gap energy, thus raising a serious question about the role of thermally activated quasiparticle tunneling for the negative magnetoresistance. Further analysis including fluctuations above H_{c2} and T_c is necessary to understand the peak effect. For a field parallel to the plane geometry, the isothermal magnetoresistance is monotonic with the field with a much larger resistive onset field. The large positive magnetoresistance at high temperatures for the $H\perp J\|$ plane is consistent with a warped cylindrical Fermi surface model as well as a two band model. Further measurements to lower temperatures and with both in-plane and out-of-plane angular control of the sample to the field are necessary to fully explore charge transport in the interlayer direction.

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