

Interlayer magnetoresistance peak in β'' -(BEDT-TTF) $_2$ SF $_5$ CH $_2$ CF $_2$ SO $_3$

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Transport measurements of interlayer magnetoresistance with the field parallel to the current direction have been performed on single crystals of organic superconductor β'' -(BEDT-TTF) $_2$ SF $_5$ CH $_2$ CF $_2$ SO $_3$. The magnetoresistance is found to display a pronounced peak as a function of magnetic field below the superconducting transition temperature, similar to those of κ -(BEDT-TTF) $_2$ X compounds. Analysis of the magnetoresistance peak in terms of a simple pair and quasiparticle tunneling model gives a physically negligible gap energy. © 1999 American Institute of Physics. [S0021-8979(99)17008-7]

Interlayer transport in layered systems has been of recent interest.¹⁻⁷ In anisotropic cuprates such as Bi $_2$ Sr $_2$ CaCu $_2$ O $_{8+x}$, interlayer resistivity exhibits a semiconducting temperature dependence while the in-plane resistivity is metallic. With the application of a magnetic 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 resistivity is typically three orders of magnitude larger than the in-plane resistivity. However, the temperature dependence is qualitatively similar for both directions, i.e., semiconducting for temperature above 100 K and metallic for temperature below it.^{8,9} Furthermore, interlayer transport in these materials has shown interesting field- and temperature-dependent magnetoresistance peak effect.¹⁰⁻¹⁴ Various models have been proposed to explain the peak effect, however, the origin still remains controversial.¹⁵

To explore the mechanism in the interlayer charge transport, we have performed transport measurement on a highly two-dimensional organic superconductor β'' -(BEDT-TTF) $_2$ SF $_5$ CH $_2$ CF $_2$ SO $_3$. Similar to other ET salts, the crystal of β'' -(BEDT-TTF) $_2$ SF $_5$ CH $_2$ CF $_2$ SO $_3$ contains conducting layers of radical cations alternating with anion layers. It is an example of an organic superconductor where both the donor layers and anion layer consist of organic molecules.¹⁶⁻¹⁸ However, unlike κ -(BEDT-TTF) $_2$ X, the resistivity of this compound shows a metallic behavior from room temperature down for both in-plane and interlayer resistivity with a resistivity anisotropic constant $\rho_{\perp}/\rho_{\parallel} \sim 1500$. It has a relatively low superconducting transition temperature of $T_c \approx 5$ K. In this article, we concentrate on the field and temperature dependence of interlayer magnetoresistance for $T < T_c$.

Single crystals of β'' -(BEDT-TTF) $_2$ SF $_5$ CH $_2$ CF $_2$ SO $_3$ were synthesized by an electrocrystallization technique described elsewhere.¹⁶ Several crystals were used in these measurements with average dimensions of $1 \times 0.78 \times 0.33$ mm. Extensive measurements were made on a crystal with $T_c = 5.4$ K. The room-temperature interlayer resistivity is about

700 Ω cm and the in-plane resistivity about 0.2 Ω cm. The interlayer resistance was measured with use of the ac 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 μ A was used to ensure linear I - V characteristics. The voltage was detected with a lock-in amplifier at low frequencies of about 312 Hz. The samples were slowly cooled down below the superconducting transition temperature. Magnetic field is applied perpendicular to conducting planes. Field sweep was performed slowly after the sample temperature has stabilized at a target temperature.

Figure 1 shows an overlay of interlayer resistivity 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, from right to left. At a fixed temperature,

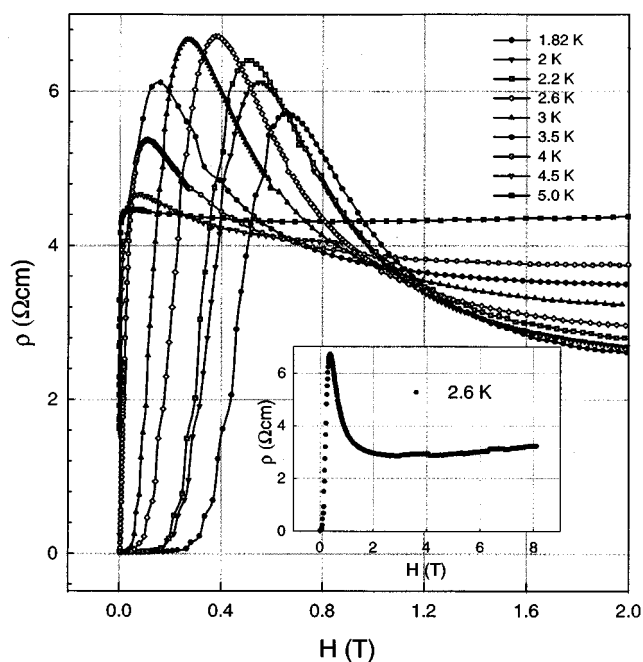


FIG. 1. Interlayer resistivity as a function of field for the H_{\perp} plane at various temperatures. The inset is a typical resistivity over a bigger field range up to 8 T at temperature 2.6 K.

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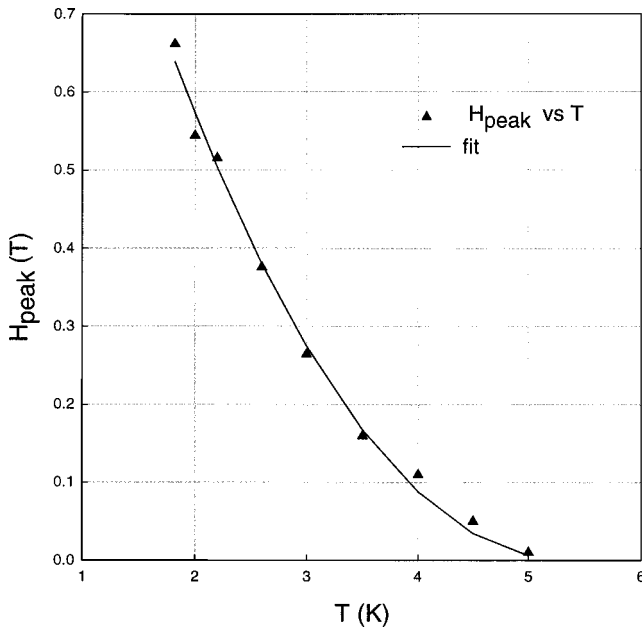


FIG. 2. Peak field vs temperature. The solid line is a fit to the data.

the resistivity becomes finite at some onset field and rises rapidly with further increase in H . At a peak field H_{peak} , the resistivity reaches a maximum ρ_{peak} . Further increase in H results in a negative magnetoresistance. With increasing temperatures, the peak field moves down toward zero. The peak effect disappears near T_c . Shown in the inset is a replot of the field dependence of resistivity as a function of field at $T = 2.6$ K over a broader field range. It is clear that resistivity increases at even higher magnetic fields.

The temperature dependence of H_{peak} is plotted in Fig. 2. The solid line is a fit to $H_{\text{peak}} = H_0(1 - T/T_c)^n$, with $H_0 = 1.5 \pm 0.2$ T, $n = 2.1 \pm 0.1$, and $T_c = 5.4 \pm 0.2$ K. The temperature dependence of the peak field is reminiscent of the irreversibility line observed in the high-temperature cuprates.

Interlayer transport for field parallel to the current direction has been studied extensively in the layered cuprates.^{4,19–21} Because the field is perpendicular to the layers, the vortex assumes the two-dimensional pancake structure. Since the current is parallel to H , there is no associated Lorentz force acting on the vortex. Even though there is a small mechanically misalignment between H and the normal of the plane, it has been argued that a small H_{\perp} plane component due to misalignment is negligible in this kind of measurement.²² In the highly anisotropic $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8-x}$ and oxygen-deficient $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the resistivity displays a peak as a function of temperature near T_c . The magnitude of the peak increases with the applied field with a corresponding decrease in the peak temperature. Several models have been proposed. For example, Dorin *et al.* have interpreted this phenomenon in terms of fluctuation theory,²³ while Gray and Kim have explained the magnetoresistance peak effect in terms of a series of stacked Josephson junctions.¹⁹

Unlike the cuprates, the layered organic superconductors display a magnetoresistance peak as a function of field at temperatures below the superconducting transition tempera-

ture in the absence of a semiconducting behavior above T_c . Based on the fact that T_c depends strongly on the applied pressure, it has been proposed that the peak effect can arise by considering the vortex–lattice interaction.¹⁰ The vortex–lattice interaction leads to local disorders in the electronic potentials, and thus, leads to an extra scattering. With increasing field, the vortices start to overlap, and eventually the large overlapping will reduce local lattice distortions and lead to the recovery of the normal transport. Although the model is plausible, there has been no structural spectroscopic evidence for the lattice distortion reported.

Another 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 magnetoresistivity. However, the presence of magnetic impurities in the all-organic $\beta''\text{-(BEDT-TTF)}_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$ 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 $\kappa\text{-(BEDT-TTF)}_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ 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. Disorders can lead to negative magnetoresistance because the magnetic field disrupts the coherent back-scattering 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.²⁴

Negative magnetoresistance has been discussed recently in terms of the stacked Josephson junction model including a field-dependent quasiparticle tunneling.^{12–14} In this model, two transport channels compete to give rise to a peak in the magnetoresistance. One channel is the Cooper pairs tunneling between the layers. For a resistively shunted Josephson junction,²⁵ the junction resistance is given by $R_t = R_{nt}[I_0(E_j/2k_B T)]^{-2}$, where I_0 is the modified Bessel function, R_{nt} the normal interlayer resistance, and E_j the Josephson coupling energy. Following the discussion by Hettinger *et al.*,²⁰ $E_j = e_j A_{\text{eff}}$, with e_j the intrinsic Josephson coupling energy, independent of the junction area. A_{eff} is the effective area which can be approximated by $A_{\text{eff}} = \Phi_0/(H + H_0)$, here Φ_0 is the flux quantum and H_0 is a fitting parameter. In the limit of $E_j \gg kT$, it can be approximated by: $R_t = R_{nt} \exp(-E_j/k_B T)$. However, at high fields, the limit of $E_j \gg kT$ is not appropriate any more and the junction resistance should be described by the full expression $R_t = R_{nt}[I_0(e_j \Phi_0/(H + H_0)k_B T)]^{-2}$. While the exact pair contribution to the conductance is not analytically calculated, it can be approximated to be proportional to $[I_0(e_j \Phi_0/(H + H_0)k_B T)]^2 - 1$, so the limiting cases are satisfied.

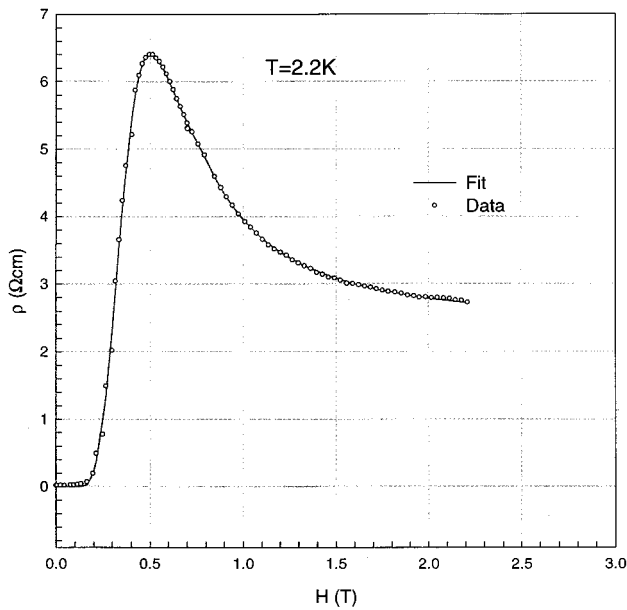


FIG. 3. Interlayer resistivity as a function of field at 2.2 K. The line is a fit to the data.

The competing channel is the tunneling of quasiparticles between the layers. If we consider the quasiparticle conductance Y_{ss} to be thermally activated $Y_{ss} \sim \exp[-\Delta(T,H)/kT]$, here $\Delta(T,H)$ is the energy gap. As discussed above, the pair conductance is $Y_p \sim [I_0(E_j/2kT)]^2 - 1$. 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 magnetoresistance.

To analyze it quantitatively, the total junction conductance is given by $Y = Y_1 \{ [I_0(e_j \Phi_0 / (H + H_0) / 2kT)]^2 - 1 \} + Y_2(H) \exp[-\Delta(T,H)/kT]$, $Y_1 = 1/R_n$. 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 assumes a constant Y_2 , we are unable to get a reasonable fit to the data. If we assume $1/Y_2(H) = R_0 [1 + (H_0/H)^n]$, a nearly perfect fit near the peak can be obtained, as shown in Fig. 3. The fit gives $1/Y_1 = 6.9 \pm 0.1 \Omega \text{ cm}$, $e_j \Phi_0 / 2kT = 0.46 \pm 0.02T$, $H_0 = -0.03 \pm 0.005T$, $1/Y_2(H) = 2.34 + 1.53/H^2 \Omega \text{ cm}$, $\Delta_0/kT = 0 \pm 0.01$, $H_{c2} = 0.9T$ for $\rho(H)$ at $T = 2.2K$. 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 $\Delta(T,H) \sim 1/H^\alpha$, the 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 the quasiparticle contribution.^{7,23} It should be

noted that in a similar approach to fit the peak effect in $\kappa - (\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$, the model failed to fit the high-field data.¹²

In summary, the interlayer magnetoresistance displays a peak effect as a function of field and temperature in the all organic superconductor $\beta'' - (\text{BEDT-TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$. The similarity with the κ -phase salts suggests the magnetoresistance peak is intrinsic to the layered systems. The magnetoresistance peak data cannot be fitted with a stacked Josephson junction model with simple thermally activated quasiparticle contributions. Inclusion of the fluctuation effect above H_{c2} may be necessary to describe the magnetoresistance peak.

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