

Interlayer magnetoresistance peak in the ET-based organic superconductors

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Abstract. We report interlayer magnetoresistance studies in the ET-based organic superconductors. For field perpendicular to the planes, the magnetoresistance displays a peak as a function of field and temperature. By studying the magnetoresistance peak as a function of superconducting transition, we find the magnetoresistance peak is intrinsic to the layered structure.

Interlayer transport in layered systems such as the highly anisotropic high temperature cuprates and the organic superconductors, is of current interest. It is important for several reasons. First, what is the role of interlayer coupling on the superconducting transition temperature [1]? Second, what is the nature of the charge transport, is it coherent or incoherent [2,3]?

The questions remain unresolved in the cuprates. Depending on the doping level, the c-axis (perpendicular to the CuO layers) transport changes from metallic to semiconducting. For example, for optimally doped cuprates, it usually displays a linear or power-law temperature dependence and it becomes nonmetallic at low temperatures for the underdoped [4]. However, for the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, even at optimal doping, the c-axis resistivity exhibits a semiconducting temperature dependence in the normal state, while the in-plane resistivity is metallic with the resistivity decreasing with decreasing temperature [6–10]. At low temperatures (below the zero field transition temperature), the interlayer magnetoresistance displays a pronounced peak as a function of temperature and field [5,11–13]. For underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, the magnetoresistance displays a pronounced peak as a function of field. Two possible models have been proposed. In one model the peak is a result of a competition between the Josephson and the quasiparticle tunneling [6,9]. The other model suggests the peak is due to a density of state fluctuation contribution to the c-axis transport [14,15]. While both models can semi-quantitatively describe the experimental results, the mechanism remains controversial.

Organic superconductors, especially the κ -(BEDT-TTF) $_2$ X [bis(ethylenedithio) tetrathiafulvalene, abbreviated as ET] family with X being $\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$, $\text{Cu}[\text{N}(\text{CN})_2]\text{Cl}$, and $\text{Cu}(\text{SCN})_2$, have shown recently physical properties very similar

to the high temperature cuprates including unconventional metallic properties [16]. The κ -(ET)₂X consists of the conducting cation layer (ET) sandwiched between the insulating anion layers (X). ET is a large planar molecule and the different possible packing patterns are denoted by different Greek letters. In the κ phase, the ET molecules form a dimer, with each dimer contributing one electron to the anions. Because of the layered structure, the κ -(ET)₂X salts have shown similar anisotropic transport properties [17,18] as the cuprates with a typical resistivity anisotropy $\frac{\rho_{\perp}}{\rho_{\parallel}} \sim 10^3$. Furthermore, the κ -(ET)₂X salts share a very similar phase diagram with the cuprates if the pressure is replaced by doping. In other words, the system can transform between unconventional metal, magnetic insulator, and a superconductor, by changing the pressure, temperature and the anion [16].

Studies of interlayer transport in the κ -(ET)₂X have revealed an interesting magnetoresistance peak as a function of field for temperatures near and below the transition temperature [19–25]. However, there are several important differences between the organic superconductors and the cuprates. One is the temperature dependence of the normal state resistance. While the cuprates typically have semi-conducting temperature dependence for the c-axis, the interlayer resistance in the organics is metallic near the transition temperature. Another major difference is the observation of a large negative magnetoresistance as a function of field in the organic compounds. Several models have been proposed to qualitatively describe the peak, such as the magnetic impurity scattering [25], vortex-lattice interaction [21], a similar mechanism as in the cuprates, involving pair and quasiparticle tunneling has been proposed by several groups [20–22]. However, a quantitative agreement between the model and the experiment is lacking [26].

We have undertaken careful studies of interlayer magnetoresistance in several compounds [21] including the κ -(ET)₂Cu[N(CN)₂]Br, κ -(ET)₂Cu(SCN)₂ salts, the all-organic β'' -(ET)₂SF₅CH₂CF₂SO₃ and the κ -(ET)₂I₃. In the κ -(ET)₂Cu[N(CN)₂]Br system, the peak has been studied as a function of transition width [23]. The peak width increases and the peak height decreases with increasing transition width. For samples with transition width greater than 2K, the peak effect disappears completely.

For high quality samples, the magnetoresistance peak was observed for all four systems which had different superconducting transition temperatures. The peak field decreases drastically with increasing anisotropy for the same reduced temperature $\frac{T}{T_c}$. The results suggest that the appearance of the peak correlates with the coupling between the layers.

Shown in figure 1 is a plot of the normalized resistive transition of four samples. The transition widths increases from about 0.2K to over 2K corresponding to sample 1 to 4. The quality of samples has drastic effect on the field dependence of the magnetoresistance at temperatures below the superconducting transition. Shown in figure 2 is an overlay of magnetoresistance as a function of field at temperatures at or close to 6K and 7K. Clearly, figure 2(a) displays sharp peak with peak field at around 3.5T and 2.5T for 6K and 7K, respectively. With increasing

transition width, the peak broadens up and disappears completely for sample 4, where magnetoresistance increases continuously with field.

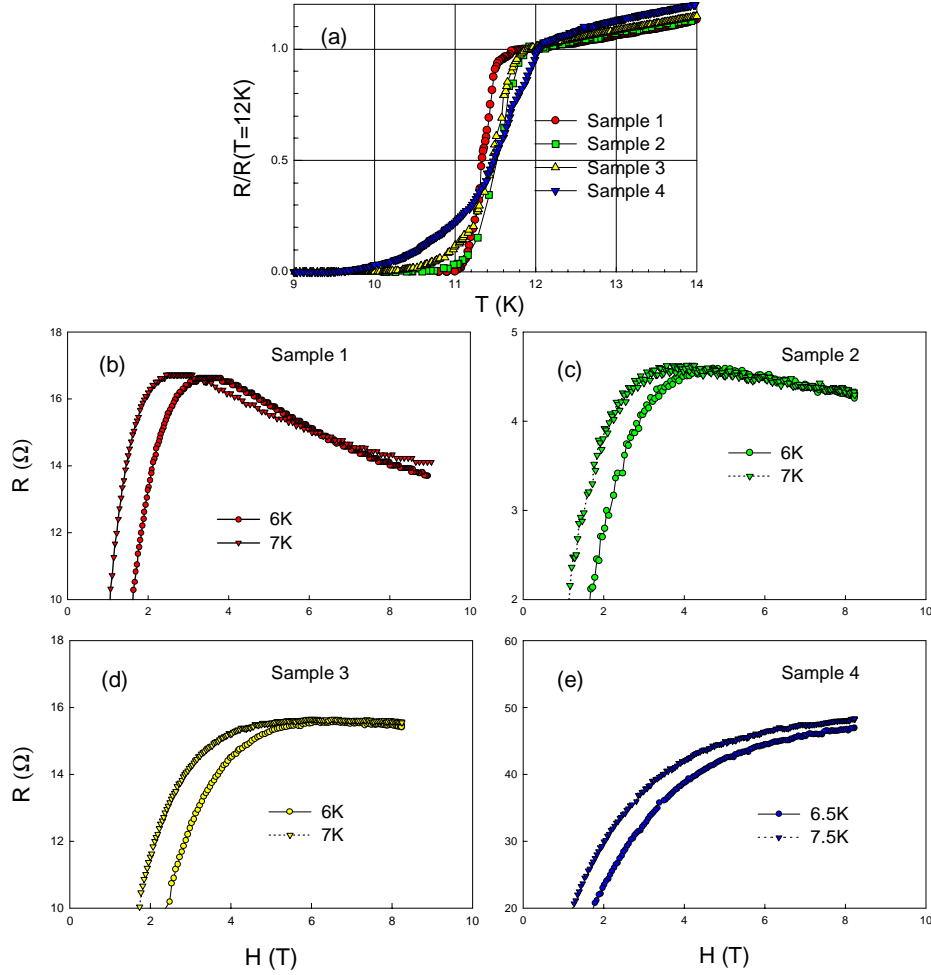


Fig. 1 (a) Normalized resistive transition for four samples. Fig. 1(b) through (e) are magnetoresistance as a function of field at different temperatures for sample 1 to 4.

The transition width dependence of the magnetoresistance peak is consistent with an earlier study where it has been shown with increasing sample quality, the interlayer peak remains while the in-plane magnetoresistance peak disappeared. The disappearance is associated with the mixing of the interlayer component in the in-plane measurements [20].

To test the magnetoresistance peak is intrinsic to the layered system, interlayer transport has been carried out in several systems with different chemical anions and superconducting transition temperatures. Shown in Figure 2(a) is a plot of the interlayer magnetoresistance as a function of field at high temperatures for an

all-organic β'' -(ET)₂SF₅CH₂CF₂SO₃. Clearly, the magnetoresistance has a pronounced peak as a function of field at temperatures below T_c. The peak field is very small compared with that of κ -(ET)₂Cu[N(CN)₂]Br. At low temperatures, as shown in figure 2(b), the magnetoresistance peak remains until 0.5K, below which the magnetoresistance is monotonic with field. It should be noted that the disappearance of the magnetoresistance peak is not unique to the β'' -(ET)₂SF₅CH₂CF₂SO₃, similar field dependence has been observed in all other systems studied at low temperatures.

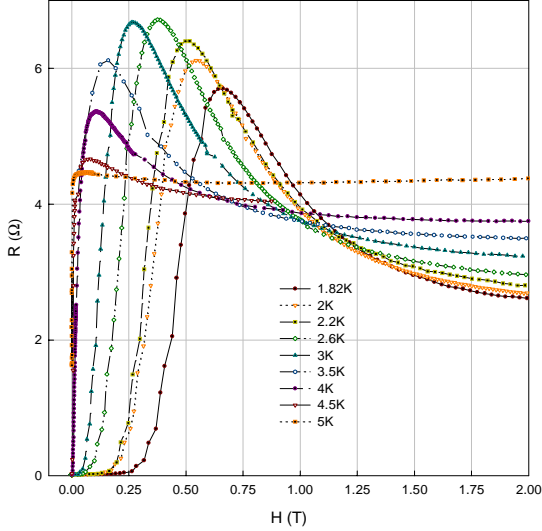


Fig. 2(a) Magnetoresistance as a function of field at high temperatures.

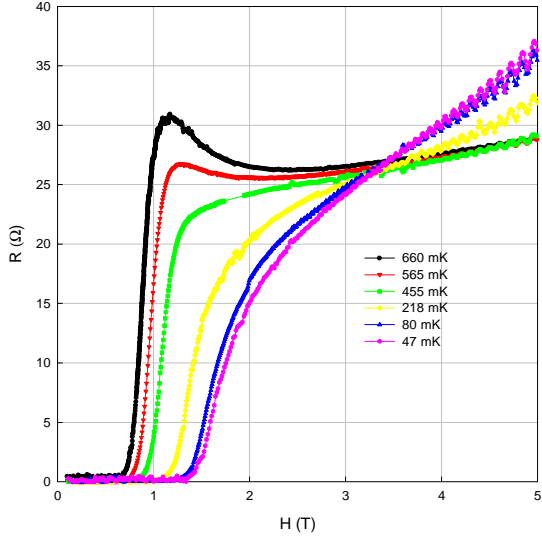


Fig. 2(b) Magnetoresistance as a function of field at low temperatures.

Figure 3 compiles the magnetoresistance peak field as a function of temperature for the four systems studied. With decreasing T_c, the H_{peak} decreases drastically. For example, the peak field for the Br salt is generally over an order of magnitude large than that of I₃ salt if compared in the reduced temperature scale. Nevertheless, the observation of the magnetoresistance peak in all systems studied suggests the universality of the field dependence of the interlayer transport in the layered compounds.

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 [14,15]. 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 vortices (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 semi-quantitatively the field and temperature dependence of the interlayer transport at $H < H_{peak}$.

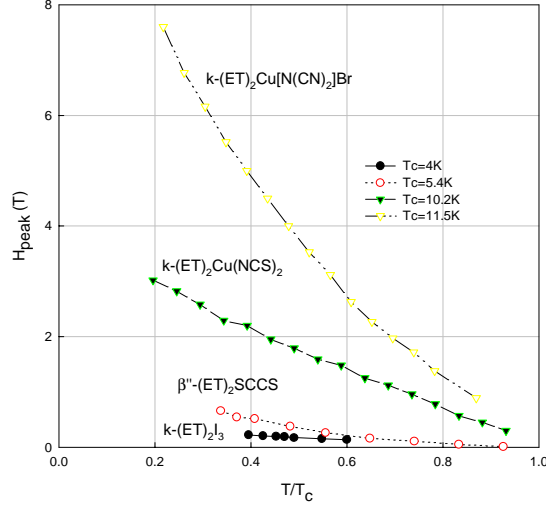


Fig. 3 Peak field versus the reduced temperature for all four samples.

In the case of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, dissipation mechanism for $H \parallel J \parallel c$ was first proposed by Briceño *et al* [6]. In this model, current moving parallel to the c axis is taken to pass through a narrow superconducting channel of area $A \approx \frac{\Phi_0}{H}$ 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's 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* [7]. 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 critical current I_c is somewhat controversial [8], a recent study

on thin mesa of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ found $I_c \propto 1/H$ [28].

A similar model has been proposed to explain the magnetoresistance peak in the organic superconductors [20,22,23]. To analyze the data quantitatively, consider the charge transport through a Josephson junction of area $a^2 \approx \frac{\Phi_o}{H+H_o}$ between the densely packed vortices, H_o being a fitting parameter to take into account of effects of pinning and lattice rigidity [9]. The junction resistance due to thermal fluctuations of the phases is given by $R(H) = R_{n'} [I_o(\frac{E_J}{2kT})]^{-2}$, where $E_J = \frac{\hbar I_c}{e} = \frac{\pi \hbar \Delta(T)}{2e^2 R_{n'}} \tanh[\frac{\Delta(T)}{2kT}]$ 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 $e_J = \frac{E_J}{a^2}$, such that $R(H) = R_{n'} [I_o(\frac{e_J \Phi_o}{(H+H_o)2kT})]^{-2}$. If $E_J \gg kT$, the junction resistance can be reduced to [9] $R(H) = R_{n'} \exp[-\frac{e_J \Phi_o}{(H+H_o)kT}]$. Shown in Figure 4 is a plot of the magnetoresistance as a function of field at various temperatures for a κ -(BEDT-TTF) $_2\text{Cu}(\text{SCN})_2$. The solid lines are fits to the stacked Josephson junction model. Clearly, the data can be reasonably well fitted over a large resistance range. The inset shows the temperature dependence of the fitted Josephson coupling energy e_J . e_J increases with decreasing temperature.

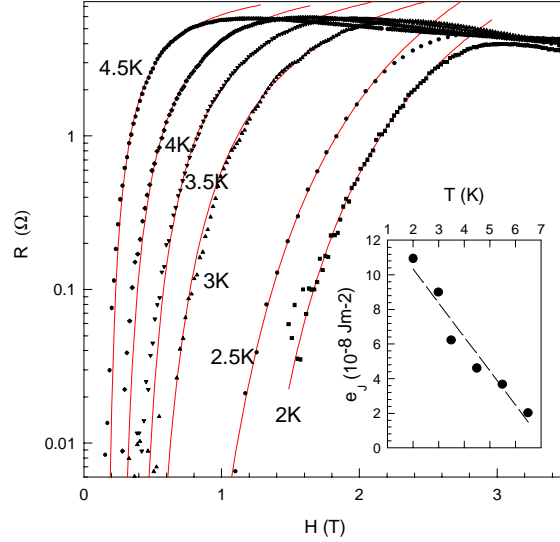


Fig. 4 Magnetoresistance as a function of field at various temperatures for SCN salt. The lines are fits to the stacked Josephson junction model. The inset show the temperature dependence of the coupling energy.

While the low field magnetoresistance can be described well with the stacked Josephson junction model, a mechanism with large negative magnetoresistance has to be invoked to explain the peak. One such model is the fluctuation contribution

to the interlayer transport near the superconducting transition. Contrary to the conventional wisdom, the positive contribution to the fluctuation conductivity in the interlayer direction is weak due to the hopping or tunneling nature of the quasiparticle propagation. Rather, the fluctuation conductivity is dominated by the negative contribution due to the decreases of density of state of quasiparticles [14,15].

The negative magnetoresistance can also arise from other interactions. For example, it has been proposed that the negative magnetoresistance in the underdoped cuprates is related to the presence of pseudogap in the oxygen deficient $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ [12]. Indeed, DC magnetic susceptibility as well as NMR relaxation measurements suggest strongly the presence of a pseudogap near 50K in the ET based compounds [29]. However, the fact that negative magnetoresistance disappears a few degrees above the superconducting transition argues against the role of pseudogap formed at much higher temperature.

Negative magnetoresistance due to the vortices in the mixed state has also been suggested. For example, thermal fluctuation of the vortices can introduce extra scattering to the charge transport. With increasing field or decreasing temperature, 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 [25]. 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, the observations of the peak in the β'' - (BEDT-TTF) $_2$ SF $_5$ CH $_2$ CF $_2$ SO $_3$ and κ -(ET) $_2$ I $_3$ where no Cu^{++} is present contradicts the model of magnetic impurities scatterings.

In summary, interlayer transport measurements on several ET based organic superconductors reveal an universal magnetoresistance peak at temperatures below the superconducting transition. At very low temperatures, the magnetoresistance peak disappears. The peak field increases with increasing superconducting transition temperature or decreasing anisotropy. The magnetoresistance peak is mostly likely due to the density of state fluctuation of quasiparticles.

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