Interlayer dissipation in magnetic fields for $H \parallel J$ in $\kappa$-(ET)$_2$I$_3$

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Abstract

In this paper, we report transport measurements of the interlayer magnetoresistance with field, $H$, parallel to the current, $J$, in the organic superconductor $\kappa$-(bis(ethylenedithio)-tetrathiafulvalene)I$_3$. The isothermal magnetoresistance $R(H)$ displays a peak as a function of field for $1 > T/T_c > 0.4$. At lower temperatures $T/T_c < 0.4$, $R(H)$ increases monotonically with field. Comparison with several other organic superconductors suggests the interlayer magnetoresistance peak is intrinsic to layered systems. Quantitative analysis shows that the magnetoresistance at small field can be fitted to the stacked Josephson-junction model with the normalized coupling energy proportional to $(1 - T/T_c)^{3/2}/H$. © 2000 Elsevier Science B.V. All rights reserved.

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Interlayer transport in highly anisotropic systems with layered structure such as the high-temperature cuprate and the organic superconductors is a topic of current interest. Depending on the doping level, the $c$-axis (perpendicular to the CuO layers) transport in the cuprates changes from metallic to semiconducting in the normal state. For example, for optimally doped cuprates, one usually observes a linear or power-law temperature dependence and a nonmetallic behavior at low temperatures for underdoped cuprates [1]. Furthermore, a Lorentz-force independent dissipation has been observed in the $c$-axis transport in magnetic field, such as in Bi$_2$Sr$_2$CaCu$_2$O$_8$ superconductors [2–6]. For underdoped YBa$_2$Cu$_3$O$_{6+x}$ and La$_2-x$Sr$_x$CuO$_4$, the magnetoresistance displays a pronounced peak as a function of field at low temperatures [7–9].

Organic superconductors, especially the $\kappa$-(BEDT-TTF)$_2$X family with BEDT-TTF =$\ldots$
bis(ethylenedithio)-tetrathiafulvalene (abbreviated as ET) and X being Cu[N(CN)₂]Br, Cu[N(CN)₂]Cl, and Cu(SCN)₂, have recently shown physical properties very similar to the high-temperature cuprates including unconventional metallic properties [10]. The κ-(ET)_2X salts consist of a conducting (ET) cation layer sandwiched between badly conducting 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 face-to-face dimers, with adjacent pairs forming almost a right angle to the neighboring ones. Each dimer contributes one electron to the anions. Because of the layered structure, the κ-(ET)_2X salts have shown similar anisotropic transport properties as the cuprates with typical resistivity anisotropies of \( \rho_{//} / \rho \simeq 10^3 \), where \( \rho_{//} \) and \( \rho \) denote the resistivity parallel and perpendicular to the ET planes, respectively [11,12].

Studies of the interlayer transport in κ-(ET)_2X have revealed an interesting magnetoresistance peak as a function of field for temperatures near and below the transition temperature [13–22]. Similar to the cuprates, large field-dependent magnetoresistances have been observed for the field parallel to the current direction where no magnetic force on the vortices is expected. Moreover, a large negative magnetoresistance has been observed in several ET-based compounds such as κ-(ET)_2Cu[N(CN)₂]Br, κ-(ET)_2Cu(SCN)₂, and β'-ET)_2SF₅CH₂CF₂SO₃. Several models have been proposed to qualitatively describe the peak, such as magnetic impurity scattering [21], vortex–lattice interaction [16,17], a stacked Josephson-junction model with competition between pair and quasiparticle tunneling between the planes. The latter has been proposed by several groups [15–18]. However, a quantitative agreement between the model and the experiment is still lacking [20].

In this paper, we report a careful interlayer-magnetoresistance measurement of the low-\( T_c \) organic superconductor κ-(ET)_2I₃ with a bulk \( T_c \) of 3.5 K. Similar to the high-\( T_c \) \( \kappa \)-phase salts, a magnetoresistance peak is observed below the superconducting transition. At very low temperatures, the magnetoresistance peak disappears. This demonstrates the magnetoresistance peak as an intrinsic property of the layered compounds. At small field, the resistive dissipation can be well fitted to a stacked Josephson-junction model with \( R = R_0(I_0(\gamma))^2 \) with the normalized coupling energy \( \gamma = \gamma_0(1 - T/T_c)^n / H \) and \( n = 1.5 \pm 0.1 \).

The investigated single crystals were prepared by the standard electrochemical process described in detail elsewhere [23]. The resistance was measured in a top-loading ¹He cryostat equipped with a superconducting magnet. High-field measurements revealed the well-known Shubnikov–de Hass oscillations commencing at about 3 T. The sample was placed with the field normal to the superconducting layers. The interlayer resistance was measured with an ac-resistance bridge with a typical excitation current of 10 \( \mu \)A.

Shown in Fig. 1 is an overlay of the interlayer magnetoresistance as a function of field for temperatures between 1.7 K and 4.12 K. The curves shift from right to left in the sequence of increasing temperature. At a fixed temperature, the resistance \( R(H) \) starts to increase rapidly from zero, going through a maximum at a peak field \( H_{\text{peak}} \). For example, at \( T = 1.7 \) K, \( H_{\text{peak}} \) is about 0.2 T. The magnetoresistance becomes positive again for \( H \) above 0.3 T. With increasing temperature, \( H_{\text{peak}} \) decreases. There is hardly any peak in \( R(H) \) at \( T = 4.12 \) K. The inset shows the resistive transition in zero field with an onset temperature at about 4.25 K.

At lower temperatures, the magnetoresistance peak disappears gradually as shown in Fig. 2. The seven curves correspond to temperatures between 1.58 and 0.6 K. At \( T = 1.28 \) K, \( R(H) \) increases monotonically, although a shoulder-like feature is still present. This feature disappears gradually with further decrease in temperature.

The magnetoresistance peak in the interlayer direction with field perpendicular to the planes has been observed in several ET-based organic superconductors. Fig. 3 compiles the temperature dependence of the peak field for four different compounds [16,17,20]. With decreasing superconducting transition temperature, \( H_{\text{peak}} \) decreases appreciably. For example, the peak field is 25 times larger in κ-(ET)_2Cu[N(CN)₂]Br than that of κ-(ET)_2I₃ at \( T/T_c = 0.4 \). Here \( T_c = 4 \) K is used, which corresponds to the midpoint in the resistive transition. The general features of the field and temperature dependence of the interlayer resistance are similar in all systems.
that show the disappearance of the magnetoresistance peak at very low temperatures.

The dissipation mechanism in the interlayer direction in the mixed state remains controversial. Because the current is applied parallel to the field direction, the usual driving Lorenz force should be absent. In the case of Bi$_2$Sr$_2$CaCu$_2$O$_{8+}$, a dissipation mechanism for $H \parallel J \parallel c$ was first proposed by Briceno et al. [2]. In this model, current moving parallel to the c axis is assumed to pass through a narrow superconducting channel of area $A = \Phi_0/H$ between the densely packed vortices. Here $\Phi_0$ is the flux quantum. Dissipation occurs through thermodynamic fluctuations that cause the phase of the superconducting order parameter in the c direction to jump by $2\pi$. With the assumption that the fluctuations in each channel are independent, the dissipation in the c direction can be modeled by a long, narrow Josephson junction at finite T [24]. The resistance of the weak link is approximately given by $R = R_n[I_c(\gamma)]^{-2}$, where $\gamma = \hbar I_c/(2ek_BT)$. $R_n$ is the normal-state resistance, $\hbar$ is Planck’s constant, $k_B$ is the Boltzmann constant, $I_c$ is the critical current, $e$ is the electron charge, and $I_0$ is the modified Bessel function. In this case, since the normal-state resistance is activated in the c direction, a resistance peak is expected 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 and Kim [3]. For an intermediate Josephson coupling, the junction conductance is the sum of the quasiparticle conductance $Y_{qs}$ and pair conductance $Y_p$, i.e., $Y = Y_{qs} + Y_p$. Assuming the quasiparticle conductance $Y_{qs}$ is thermally activated $Y_{qs} \propto \exp[-\Delta(T, H)/k_B T]$, and the pair conductance $Y_p \propto [I_c(y/\gamma)]^2 - 1$, a distinct peak in $R(T)$ arises naturally. However, the field dependence of the critical current $I_c$ is somewhat controversial [4]. In a recent study on a thin mesa structure of a Bi$_2$Sr$_2$CaCu$_2$O$_8$ crystal, $I_c \propto 1/H$ was found [25].

This model can be applied to the dissipations observed for the organic superconductors [15,18,19]. To analyze the data quantitatively, we consider the charge transport through a Josephson junction of area $a^2 \equiv (\Phi_0)/(H)$ between the densely packed vortices [5]. Because $I_c$ is proportional to the junction area $I_c = J_c a^2$, it can be reexpressed as $hJ_c \Phi_0/2e k_B T H$. For large $\gamma$ the junction resistance can be reduced to $R = R_o \exp[-(hJ_c \Phi_0/2e k_B T H)]$. In order to check the validity of this model, the magnetoresistance is plotted as a function of inverse field at several temperatures in a semi-log scale for $\kappa$-(ET)$_2$I$_3$ in Fig. 4. Indeed, for a resistance below about 0.1 $\Omega$, $R(H)$ decreases exponentially with $1/H$. The solid lines are fits to the exponential expression for large argument. While the field range is relatively small, the resistance data can be reasonably well fitted over about three orders of magnitude. The magnitude of the slope increases with decreasing temperature.

To see the temperature dependence of the slope, Fig. 5 shows $\mathrm{d} \log R/\mathrm{d}(1/H)$ vs. $T$. The solid line
Fig. 3. Peak field as a function of reduced temperature for several ET-based superconductors.

is a fit to the expression $\gamma(H) = \gamma_0(1 - T/T_c)^n/H$ with $T_c = 3.3 \pm 0.1$ K, $n = 1.5 \pm 0.1$ and $\gamma_0 = 4.9 \pm 0.2$ T. Within experimental errors, the curve reveals the reasonable fit to the data. Note the $T_r$ fitted from $\gamma(H)$ is close to the zero-resistance temperature, not the resistive onset temperature. To check whether the parameters are physical, we estimate the critical

current by use of $J_c = (ek_b T/h \Phi_0) \gamma$. At $T = 1$ K, we obtain $J_c \approx 5 \times 10^3$ A/cm$^2$. The low critical current is consistent with the overall very anisotropic nature of this organic superconductor [26,27]. While there is no direct $J_c$ measured in this compound, we note that the value extrapolated above is considerably larger than the $J_c$ measured in $\kappa$-(ET)$_2$Cu(SCN)$_2$ [18]. The difference may be due to the fact that we define the $J_c$ in terms of the effective areas between the vortices, rather than the total area of the crystal.

The field and temperature dependence of the normalized Josephson-coupling energy $\gamma(H)$ is very similar to that of the irreversibility line and the resistive-transition width in the cuprates [28,29]. For highly anisotropic systems, the irreversibility-field line $H_{irr}(T)$ is usually found to display a $(1 - T/T_c)^n$ temperature dependence with $n = 3/2$ for $T/T_c \geq 0.6$. The irreversibility line has often been associated with the dimensional crossover of the vortices or the vortex-melting line. The temperature dependence can be obtained by using the heuristic argument that the activation energy associated with the flux motion has the form $U_c \propto H_c^2 V$, where $H_c$ is the thermodynamic critical field and $V$ is a characteristic excitation volume. Different models exist to estimate $V$. For $\kappa$-(ET)$_2$I$_3$, the best description of $H_{irr}(T)$ deter-

Fig. 4. Magnetoresistance as a function of inverse field in a semi-log scale at several temperatures. The lines are fits to the stacked Josephson junction model.

Fig. 5. $-d(\log R)/d(1/H)$ as a function of temperature. The solid line is a fit.
mined by a.c.-susceptibility measurements [26] is obtained by assuming $V \propto a_0^2$, where $a_0 \propto (\Phi_0/H)^1$ is the flux-line spacing in a field $H$. With $H_\text{ir}(T) \propto (1 - T/T_c)$ this results in $H_\text{ir}(T) \propto (1 - T/T_c)^{\nu/2}$. For $V \propto a_0^2 \xi$ with the coherence length $\xi \propto (1 - T/T_c)^{\nu/2}$ one obtains $H_\text{ir}(T) \propto (1 - T/T_c)^{\nu/2}$. Indeed, the $H_\text{ir}$ data of [26] might be equally well fitted by the latter temperature dependence. Similar arguments may be implied for the interlayer dissipation due to thermal fluctuations of vortices. The present work shows that $\gamma(H) = \gamma_0(1 - T/T_c)^{\nu/2}/H$ holds for the entire temperature range measured (down to $T/T_c \sim 0.2$) in good agreement with the a.c.-susceptibility data [26,30].

A $1/H$ dependence in critical current has also been inferred from Josephson-plasma resonance measurements of Bi$_2$Sr$_2$CaCu$_2$O$_8$ [31–33]. It has recently been shown that the critical current $J_c(H,T) = J_c(0,T) \cos \varphi_{n+1} \propto H^{-\mu}$ with $\mu = 1 \pm 0.1$, where $\cos \varphi_{n+1}$ is the interlayer gauge-invariant phase difference averaged thermally and spatially [31]. Direct transport measurements in a well-defined thin mesa sample of Bi$_2$Sr$_2$CaCu$_2$O$_8$ confirm the $1/H$ dependence of the critical current. In the case of organic superconductors, Josephson-plasma resonance has indeed been observed in $\kappa$-(ET)$_2$Cu(SCN)$_2$-Br [34]. However, the exponent is found to be around 0.7 $\pm$ 0.1%. The different $\mu$ values were attributed to different pinning mechanisms in the two systems. Josephson-plasma resonance studies in the present compound would be highly desirable to verify the exponent.

While the low-field magnetoresistance seems to be consistent with the stacked Josephson-junction model, a mechanism with large negative magnetoresistance has to be invoked to explain the peak. We examine several possibilities in the following: (a) A negative magnetoresistance due to the presence of a pseudogap. For example, it has been proposed that the negative magnetoresistance in the underdoped cuprates is related to the presence of a pseudogap in oxygen-deficient YBa$_2$Cu$_3$O$_{6+x}$ [8]. Indeed, d.c. magnetic susceptibility as well as NMR relaxation measurements suggest the presence of a pseudogap near 50 K in the ET-based compounds [35]. However, the fact that the negative magnetoresistance disappears a few degrees above the superconducting transition argues against an origin due to a pseudogap formed at much higher temperatures. (b) The effect of magnetic impurities. It has been suggested in a study of $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Br that traces of Cu$^{2+}$ ions might be sufficient for the negative magnetoresistance [21]. With increasing field, the effect of magnetic scattering is reduced. However, the absence of a negative magnetoresistance in the normal state $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Br suggests against the magnetic-impurity model. The present study adds another piece of evidence since there is no Cu$^{2+}$ present in $\kappa$-(ET)$_2$I$_3$. Similar conclusion has been drawn from the observations of the peak in the organic superconductor $\beta$-$\kappa$-(ET)$_2$SF$_6$CH$_2$CF$_2$SO$_3$ [20]. (c) Fluctuations of the quasiparticle density of state. In this case, the positive contribution to the fluctuation conductivity in the interlayer direction is weak due to the hopping or tunneling nature of the quasiparticle propagation. The fluctuation conductivity is instead dominated by the negative contribution due to the decreases of density of state of quasiparticles [36–38]. The magnetoresistance peak arises from the competition between the stacked Josephson-junction contribution with the enhanced quasiparticle tunneling at high field. However, quantitative analysis is necessary to test the fluctuation model.

In summary, we have observed a systematic evolution of the interlayer-magnetoresistance peak as a function of temperature in $\kappa$-(ET)$_2$I$_3$. At high temperatures, $T/T_c$ $\geq$ 0.4, a peak in $R(H)$ is clearly observed with the peak-field increasing with decreasing temperature. For $T/T_c$ $\leq$ 0.4, $R(H)$ increases monotonically with field. Comparison with several other ET-based organic superconductors suggests that the peak field is reduced drastically for samples with lower $T_c$ or higher anisotropies. Quantitative analysis of the low-field magnetoresistance shows that $R(H)$ can be described by a stacked Josephson-junction model with $\gamma(H) = \gamma_0(1 - T/T_c)^{\nu}/H$. The negative magnetoresistance may arise from fluctuations of the quasiparticle density of state.

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