Temperature Dependence of Inter-Layer Longitudinal Magnetoresistance in \(\alpha-(BEDT-TTF)_{2}\)I\(_2\): Positive versus Negative Contributions in a Tilted Dirac Cone System

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Temperature dependence of the longitudinal interlayer magnetoresistance (MR) of a bulk Dirac cone system, \(\alpha-(BEDT-TTF)_{2}\)I\(_2\), is examined. It exhibits a peak related to the crossover from the quantum limit regime with negative MR at low temperatures to the inter-level scattering regime with positive MR at higher temperatures. The field dependence of the MR peak temperature, which measures the energy separation between the \(n = 0\) and \(\pm 1\) Landau levels, evidences that the peculiar Landau quantization of the Dirac cone dispersion is relevant to both effects. The effective Fermi velocity, \(v_{\text{eff}}\), which reflects the width and spacing of the Landau levels, has been estimated from the analysis taking account of the Zeeman-split Landau levels.

KEYWORDS: \(\alpha-(BEDT-TTF)_{2}\)I\(_2\), Dirac fermion, Landau level, magnetoresistance, inter-layer resistance

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A layered organic conductor \(\alpha-(BEDT-TTF)_{2}\)I\(_2\) (BEDT-TTF = bis(ethylenedithio)-tetrathiafulvalene) is known to behave as a two-dimensional massless Dirac fermion system under high pressures. Under ambient pressure, this material is a gapped insulator with charge ordering below about 135 K.\(^{1,2}\) Pressure suppresses the charge ordering so as to close the gap.\(^3\)–\(^7\) The band structure of this material under pressure is characterized by a linear energy dispersion,\(^6\)–\(^7\) expressed as \(E = \pm v_0 \sqrt{k^2 + \epsilon^2} / C_{30}\) where plus and minus signs mean electron and hole bands respectively, \(k = |k|\) is the absolute value of in-plane wave vector and \(v_0\) is the anisotropic Fermi velocity depending on the azimuth angle \(\phi\) of \(k\). The Fermi energy is set to zero where the electron and hole bands contact together. This Dirac cone type linear dispersion is found around two points in the first Brillouin zone. This peculiar energy spectrum causes anomalous transport phenomena, such as a temperature independent in-plane resistance or a carrier density obeying a power law of temperature, for example.\(^4\)–\(^9\) Unlike the well-known Dirac cone system graphene\(^{10}\) (monolayered graphite), \(\alpha-(BEDT-TTF)_{2}\)I\(_2\) is a bulk system and the cones are tilted due to low crystal symmetry.\(^7\) The tilted feature is expressed by the \(\phi\)-dependent Fermi velocity.

One of characteristics of the Dirac cone is its peculiar Landau quantization. When magnetic field, \(B\), normal to the conducting plane is applied, the linear dispersion is quantized to discrete Landau levels, \(E_n = \pm v_0 \sqrt{2\epsilon\hbar B / C_{30}}\), where \(v_0\) is effective Fermi velocity with the tilting effect, \(n\) is the Landau index, and positive and negative signs correspond to electron and hole levels respectively. It is characterized by the \(n = 0\) level at Fermi energy and by a large energy separation, \(\Delta E_0 = v_0 \sqrt{2\epsilon\hbar B} / C_{30}\), proportional to \(\sqrt{B}\) between the \(n = 0\) and \(\pm 1\) levels. Because of the large \(\Delta E_0\), the quantum limit is easily approached in this system, so as to cause anomalous MR effects. For example, strong negative inter-layer MR is observed at low temperatures under strong magnetic fields.\(^{11}\) Osada\(^{12}\) and Morinari \textit{et al.}\(^{13}\) have explained the negative MR in terms of the Landau quantization. Since each Landau level has degeneracies proportional to \(B\), density of state at the each level becomes very high under strong magnetic fields. Therefore, the inter-layer conductivity proportional to the density of state at Fermi energy (the \(n = 0\) level) appreciably enhances the quantum limit. This gives rise to the negative MR as found in our experiments for \(\alpha-(BEDT-TTF)_{2}\)I\(_2\). This finding has clearly evidenced the Dirac cone dispersion in \(\alpha-(BEDT-TTF)_{2}\)I\(_2\).

On the other hand, at higher temperatures, positive longitudinal MR is observed (Fig. 1).\(^{11}\) This positive contribution appears even at about 50 K, and remains at low fields at low temperatures, where the negative MR is dominant at high fields. Since the effect of the Lorentz force should be small in the longitudinal configuration (the current is nearly parallel to the field direction), this result suggests a mechanism related to the large \(\Delta E_0\). In the transverse measurements \((B \perp \text{layer})\), positive MR similarly appears even at high temperatures up to about 50 K.\(^{4}\) The similarity of the temperature dependence of the longitudinal and transverse resistivity under various fields suggests a possible relation between the origins of these two phenomena. From the quadratic field dependence of the transverse MR at low fields, it has been regarded as a result of the Lorentz force operating on the carriers with very high mobility in terms of the semiclassical picture. It has pointed out recently that the MR can be related to the interaction between the contact points.\(^9,14\) For the inter-layer resistance, Morinari and Tohyama have recently explained its origin in terms of the Landau quantized two-dimensional system weakly interacting with tilted inter-layer transfer integrals.\(^{15}\) The inter-layer transfers, whose tilting is necessary for the longitudinal MR,
give rise to transitions between the Landau levels when the electrons move from layer to layer.\cite{13} The transitions are suppressed in the quantum limit, but are promoted by the broadening of the Landau levels at high temperatures to yield positive MR under field. The competition between the positive and negative MR effects provides a peak in the field dependence of resistance. The relevance of the Landau quantization theory\cite{15} to the observed field dependence of resistance has been discussed previously.\cite{11}

In this paper, we focus on the temperature dependence of MR. The MR exhibits a crossover peak, as well as in the field dependence data. We show the details of the inter-layer MR behavior of \textit{\alpha}-\textit{(BEDT-TTF)}$_2$I$_3$. The field dependence of the peak temperature is shown to be expressed mainly in terms of the Landau quantization effect. From the analysis the effective Fermi velocity value is deduced.

We performed MR measurements using single crystals of \textit{\alpha}-\textit{(BEDT-TTF)}$_2$I$_3$ prepared by an electrochemical method. Hydrostatic pressures were applied on the sample by using a pressure cell made by CuBe. Four Au wires were attached on the sample surface, and a conventional DC method was adopted for resistance measurement. Both electrical current and magnetic field were applied in the direction normal to the conducting plane. We measured temperature dependence of the inter-layer resistance under the longitudinal field with fixed field strength. Experiments were performed below 100 K for several magnetic fields up to 7 T under 1.5 GPa.

Below 100 K, the inter-layer resistance \textit{R}$_{zz}$ at \textit{B} = 0 shows a similar temperature dependence to the in-plane resistance. \textit{R}$_{zz}$ is nearly constant for temperature showing the slightly metallic behavior down to around 15 K where it exhibits the minimum. It shows upturn below 15 K.

Anomalous temperature dependence of \textit{R}$_{zz}$ is observed under field. Figure 1 shows the temperature sweep of \textit{R}$_{zz}$ under 5 T, where the background (the \textit{B} = 0 data) is also shown. The ordinary orbital MR due to the Lorentz force should not be important under the longitudinal field. Nevertheless MR, the deviation from the background, appears distinctly below about 50 K. This MR is not very large in this temperature region. However, there is clearly a peak of interest at \textit{T}$_{\text{max}}$ = 14 K at \textit{B} = 5 T.

The MR data are normalized as \textit{M}$_R$ = \textit{R}$_{zz}$(\textit{B})/\textit{R}$_{zz}$(\textit{B} = 0) − 1. The normalized data are shown in Fig. 2. The MR peak is highlighted in this figure. The MR is positive in the wide temperature region except for the lowest temperatures where the negative MR is dominant. The positive MR appears below around 50 K. Making the peak at \textit{T}$_{\text{max}}$, the MR rapidly increases above \textit{T}$_{\text{max}}$, but drops below \textit{T}$_{\text{max}}$ with lowering the temperature. The crossover from the positive to negative MR occurs at \textit{T}$_{\text{max}}$, a boundary between the positive and negative MR regimes. Since the negative MR is observed below \textit{T}$_{\text{max}}$, it is clear that the quantum limit is nearly approached below \textit{T}$_{\text{max}}$. The positive and negative contributions thus compete with each other.

We also measured the field dependence of \textit{M}$_R$ vs \textit{T} curve. The curve changes quantitatively with varying field. Several temperature sweeps of the MR at various fields are shown in Fig. 3. The peak of the MR is observed at all fields, even at the lowest field 0.25 T. The peak height enhances accompanied by the positive shift of \textit{T}$_{\text{max}}$ as the field increases.

For the field dependence, the appearance of the MR peak\cite{11} has been explained by Morinari and Tohyama in terms of the Landau quantization. According to their theory, the effective energy separation between the \textit{n} = 0 and ±1 levels, as a function of \textit{B}, approximately equals to the width of the level, \textit{T} = \textit{k}_{\text{B}}\textit{T}$^\prime$, at the MR peak field. We surmise here a similar condition holds at the MR peak in the temperature dependence. The MR data shown in Figs. 2 and 3 are then interpreted as follows: At sufficiently high temperatures above \textit{T}$_{\text{max}}$, the thermal broadening of the Landau levels becomes much larger than the level separations. Therefore, the MR is zero at \textit{T}$\gg$\textit{T}$_{\text{max}}$, because the Landau quantization is diminished. The broadened levels with the large overlap gradually grows with lowering temperature, then, the positive MR increases due to the inter Landau level scattering. The scattering turns to

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**Fig. 1.** (Color online) Temperature dependence of inter-layer resistance (\textit{R}$_{zz}$) at \textit{B} = 5 T under hydrostatic pressure \textit{P} = 1.5 GPa in \textit{\alpha}-(BEDT-TTF)$_2$I$_3$. The background resistance at \textit{B} = 0 T is also shown. The arrow indicates the MR peak temperature \textit{T}$_{\text{max}}$.

**Fig. 2.** (Color online) Temperature dependence of MR (\textit{M}$_R$) obtained from the \textit{R}$_{zz}$ data in Fig. 1. \textit{M}$_R$ = \textit{R}$_{zz}$(\textit{B})/\textit{R}$_{zz}$(\textit{B} = 0) − 1.
The levels decrease. The scattering between the decrease at lower temperatures, since the thermal overlaps of the Landau levels have offset width $\Gamma$ due to the carrier scattering by charged defects. According to the self-consistent Born approximation, $\Gamma = a v / \sqrt{2 e B \hbar}$, where $a = n_{i} \alpha^{2} / (2 \pi v^{2} \hbar^{2}) < 1$, $n_{i}$ is the impurity density per unit area, and $u$ is the strength of the impurity potential per unit area. The Zeeman effect is taken into account by the last term in eq. (1). Similarly to our previous study, we set $g = 2$ here. The Zeeman (spin) splitting of the Landau levels also reduces the inter-level separation. The least squares fit to $T_{\text{max}} = c \Delta E_{\text{net}} / k_{B}$ is shown by the solid curve in Fig. 4.

The reproducibility of the data is satisfactory. We obtain, $c = 0.93$ and $v_{\text{eff}} = 2.4 \times 10^{4} \text{ m/s}$ from the fitting. To check the validity of the analysis, we compare the results with those for the field dependence.

From the behavior of the peak field $B_{p}$, a parameter $C = T_{B}^{1/2} \approx 10 \text{ K} T^{-1/2}$ has been estimated from the experimental data for $\alpha$-(BEDT-TTF)$_{2}$I$_{3}$. In our analysis, $C = v_{\text{eff}} / \sqrt{2 e B / \hbar}$, which is calculated to be $9.3 \text{ K} T^{-1/2}$. These values of $C$ well agree with each other. The temperature and field dependence of the MR peak is thus consistently analyzed, so that our present scheme in terms of the Landau quantization is justified from the experimental viewpoint. The level separation parameter, $\Delta E_{01}$, for the present data is also obtained as a function of $B$, i.e., $\Delta E_{01} / k_{B} = C / B$. The peculiar $n \approx 0$ and $\pm 1$ Landau level structure is thus revealed for the first time for $\alpha$-(BEDT-TTF)$_{2}$I$_{3}$ by its temperature dependence of MR.

In the present data, the effect of the scattering due to charged defects is not negligible. In fact, we obtain $\alpha = 0.76$ using the averaged Fermi velocity $v = 1 \times 10^{4} \text{ m/s}$ obtained from the Hall coefficient, so that we have $v_{\text{eff}} = 0.24 v$. It is possible to estimate the offset width at an appropriate field by the relation, $\Gamma = a v / \sqrt{2 e B \hbar}$. For $B_{p} \approx 0.2 \text{ T}$ at low temperatures, $\Gamma / k_{B} \approx 14 \text{ K}$ is obtained. This value is larger than $\Gamma / k_{B} \approx 3 \text{ K}$ estimated for our previous measurements on a different sample. For this material, insufficient pressure remains charge ordered defects. Strong anisotropy in $v_{H}$ may be relevant for this discrepancy. We have used the $v$ value obtained from the in-plane Hall measurements, which may be overestimated for the average $v$ value to give Landau quantization. If a smaller value, $v = 0.5 \times 10^{4} \text{ m/s}$, for example, is assumed for the level separation, rather than that used above, a more appropriate estimation $\Gamma / k_{B} \approx 3.5 \text{ K}$ is obtained. Another possibility is that the inter-layer transfers accompanied by inter-level transfers also broaden the Landau levels, which may contribute to the offset width $\Gamma$. Even if these factors are taken account of, the discrepancy is so large that the defect scattering is significant in the present case. For more detailed discussion on the sizable difference between $v_{\text{eff}}$ and $v$, sample and pressure dependence should be examined. It is not easy to derive relevant information from the height of the MR peak, because the height is determined by the balance between the positive and negative contributions.

$$k_{B} T_{\text{max}} = c \Delta E_{\text{net}}$$
$$= c (v \sqrt{2 e B} - g \mu_{B} B - \Gamma)$$
$$= c v_{\text{eff}} \sqrt{2 e B} - c g \mu_{B} B$$,

(1) where $v_{\text{eff}} = v (1 - \alpha)$ is modified effective Fermi velocity, $g$ is the $g$-factor, and $c$ is a dimensionless coefficient of order of unity. Here, $v_{\text{eff}} (< v)$ is used to take account of the fact that the Landau levels have offset width $\Gamma$ due to the carrier scattering by charged defects.
In summary, the relevance of the Landau quantization to the temperature dependence of MR in $\alpha$-(BEDT-TTF)$_2$I$_3$ has been shown. On the basis of the peculiar energy structure of the Dirac cone system under field, the MR peak temperature has been shown to give information on the energy separation of the Landau levels. The unique characteristics of $\alpha$-(BEDT-TTF)$_2$I$_3$, i.e., the weakly coupled layers having the tilted Dirac cone dispersion, are responsible for the appearance of positive MR at higher temperatures. The effective Fermi velocity, $v_{\text{eff}}$, is introduced to explain the observed behavior of the MR peak. The small value of the estimated $v_{\text{eff}}$ indicates some extra origin of the broadening of the Landau levels.

Acknowledgments

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