# Third Angular Effect of Magnetoresistance in Quasi-One-Dimensional Conductors

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We theoretically and experimentally study a new type of angular effect of magnetoresistance in quasi-one-dimensional (Q1D) conductors. It is a kink structure on the angular dependence of interlayer magnetoresistance when the magnetic field is rotated in the most conducting plane. This effect originates from the appearance or vanishing of closed orbits on the sheetlike Fermi surface. This is "the third angular effect" in the Q1D conductors following the Lebed resonance and the Danner-Chaikin oscillation. [S0031-9007(96)01983-7]

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In the past several years two novel angular effects of magnetoresistance (MR) have been discovered in the quasi-one-dimensional (Q1D) conductors having a pair of sheetlike Fermi surfaces (FS's). The first angular effect, which is called the Lebed resonance or the magic angle effect, is a series of resonancelike dip structures on the angular dependence pattern of MR when the magnetic field is rotated in the plane perpendicular to the one-dimensional (1D) axis [1–3]. The second angular effect, called the Danner-Chaikin oscillation, is the oscillation of MR when the magnetic field is rotated in the plane defined by the 1D axis and the normal of the most conducting plane [4]. These two phenomena relate to the delicate warping features of the sheetlike FS's. Basically they can be understood in the framework of the semiclassical magnetotransport theory [4–6]. Experimentally they have been studied mainly in organic Q1D conductors $(\mathrm{TMTSF})_2X$, where TMTSF denotes tetramethyltetraselenafulvalene and $X = \mathrm{ClO}_4$, $\mathrm{PF}_6$, and $\mathrm{NO}_3$.

Recently, Yoshino *et al.* observed a different kind of angular effect in another organic Q1D conductor $(\mathrm{DMET})_2\mathrm{I}_3$, where DMET denotes dimethyl(ethylenedithio)diselenadithiafulvalene [7]. It was a single kink structure on the angular dependence pattern of MR when the magnetic field was rotated in the "third" direction, that is, in the most conducting plane. They ascribed this phenomenon to some change of the electronic state. Nevertheless its origin has still been unclear because the electronic structure of $(\mathrm{DMET})_2\mathrm{I}_3$ has not been established yet. In addition, the relation between the newly found angular effect and the conventional ones, the Lebed resonance and the Danner-Chaikin oscillation, has been also unclear since neither of the conventional effects has been reported in $(\mathrm{DMET})_2\mathrm{I}_3$.

In this paper we theoretically and experimentally study a possible new type of FS topological effect of the Q1D conductors, say, "the third angular effect." It is considered to be the most plausible mechanism for the new phenomenon found in $(\mathrm{DMET})_2\mathrm{I}_3$. By the numerical calculation, we see that the semiclassical magnetotransport theory leads to the third angular effect in the same way as the Lebed resonance and the Danner-Chaikin oscillation. To confirm this numerical prediction, we experimentally search for the third angular effect in the most established Q1D organic conductor $(\mathrm{TMTSF})_2\mathrm{ClO}_4$, in which the other two conventional effects have been studied well.

First, we generally consider the dependence of MR on the magnetic field orientation in the Q1D conductor having a pair of sheetlike FS's. We take the following tight-binding band model:

$$E(\mathbf{k}) = -2t_a \cos ak_x - 2t_b \cos bk_y - 2t_c \cos ck_z - E_F \,. \tag{1}$$

Here, $a$, $b$, and $c$ are the lattice constants and $t_a$, $t_b$, and $t_c$ are the transfer integrals along the $x$, $y$, and $z$ axis, respectively. We introduce large anisotropy into the system by setting $t_a \gg t_b \gg t_c$. In this case the $x$ axis and the $x$-$y$ plane correspond to the conducting 1D axis and the most conducting plane (2D layer), respectively. We locate the Fermi level $E_F$ in the range $-2t_a + 2t_b + 2t_c < E_F < 0$, so that the system has a pair of sheetlike FS's perpendicular to the $k_x$ axis in the $\mathbf{k}$ space.

In the semiclassical picture, the electron orbital motion under magnetic fields is described by the following equations of motion:

$$\hbar\dot{\mathbf{k}} = (-e)\mathbf{v} \times \mathbf{B}, \qquad \mathbf{v} = (1/\hbar)[\partial E(\mathbf{k})/\partial \mathbf{k}]. \tag{2}$$

The conductivity tensor elements can be calculated from the electron orbital motion using the kinetic form of the

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Boltzmann equation:

$$\sigma_{ij} = \frac{2e^2}{V} \sum_{\mathbf{k}} \left( -\frac{df}{dE} \right) \nu_i(\mathbf{k}, 0) \int_{-\infty}^{0} \nu_j(\mathbf{k}, t) e^{t/\tau} \, dt \,. \quad (3)$$

Here the relaxation time $\tau$ is assumed as a constant (the relaxation time approximation). The resistivity tensor is obtained as the inverse of the conductivity tensor.

By estimating the above formulae we carried out the numerical simulation of the angular effects of MR in the Q1D system. We set $t_a : t_b : t_c = 1 : 0.2 : 0.05$, $a = b = c$, and $E_F = -\sqrt{2} t_a$ in the present calculation. The value of $E_F$ corresponds to the quarter filling of the Q1D band. Figure 1 shows the calculated angular dependence of the diagonal resistivity elements $\rho_{yy}$ and $\rho_{zz}$. In these diagrams the direction and the distance from the origin indicate the field orientation and the resistivity value in logarithmic scale, respectively. The resistivity is normalized by the factor $\pi abcB/2e$. The interchain resistivity $\rho_{yy}$ in the conducting layer shows monotonous angular dependence with no clear structure as seen in Fig. 1(a). In contrast, the interlayer resistivity $\rho_{zz}$ shows rich angular dependent features as shown in Fig. 1(b). The Lebed resonances, which appear when the field is rotated in the *y*-*z* plane, are successfully reproduced on the *y*-*z* plane in Fig. 1(b). The Danner-Chaikin oscillations, which appear when the field is rotated in the *x*-*z* plane, are also reproduced on the *x*-*z* plane in Fig. 1(b).

In addition to these conventional angular effects we can clearly see a sharp kink structure on the *x*-*y* plane in Fig. 1(b). We call this newly found angular effect the third angular effect. This effect causes a single kink structure in the angular dependence of the interlayer resistivity when the magnetic field is rotated in the most conducting plane. Essentially the third angular effect is semiclassical FS topological effect in the Q1D system, since it is lead by the semiclassical magnetotransport theory in the same way as the Lebed resonance and the Danner-Chaikin oscillation. The dependence of the third angular effect on the magnetic field strength and the relaxation time can be seen in Fig. 2. The kink structure becomes sharper as increasing the parameter $\tau/(\hbar^2/2t_a a^2 eB)$, which is proportional to the field strength and the relaxation time.

As is well known, the large anisotropy in the *y*-*z* plane ($t_b \gg t_c$) is very important for clear appearance of the conventional angular effects [5,6]. It was found that this anisotropy is also important for the third angular effect. When the Q1D system has large anisotropy in the plane perpendicular to the 1D axis, the interlayer resistivity clearly shows these three types of angular effects.

We should mention the 1D axis resistivity $\rho_{xx}$ measured in most experiments. In the framework of the relaxation time approximation, $\rho_{xx}$ shows no magnetic field dependence. Therefore, in order to reproduce the observed features of $\rho_{xx}$, we have to study the scattering mechanism beyond the relaxation time approximation used in the present work.

Next we consider the physical origin of the third angular effect. We check the possible FS topological effects by studying the **k**-space orbits on the FS's. When the magnetic field is rotated in the conducting *x*-*y* plane, the electron orbits are usually winding and open along the $k_z$ direction. The winding amplitude of these open orbits does not exceed the Brillouin zone width $2\pi/b$. Therefore, in the present configuration we cannot expect the Danner-Chaikin oscillations because it is necessary for the Danner-Chaikin effect that the open orbits are winding over plural zones [4]. This fact shows that the third angular effect is a perfectly different phenomenon from the Danner-Chaikin effect.

When the field angle $\theta$ measured from the 1D axis is small enough, there also exist the closed cyclotron orbits on the FS's as shown in the inset of Fig. 2. As increasing the field angle $\theta$, these closed orbits decrease

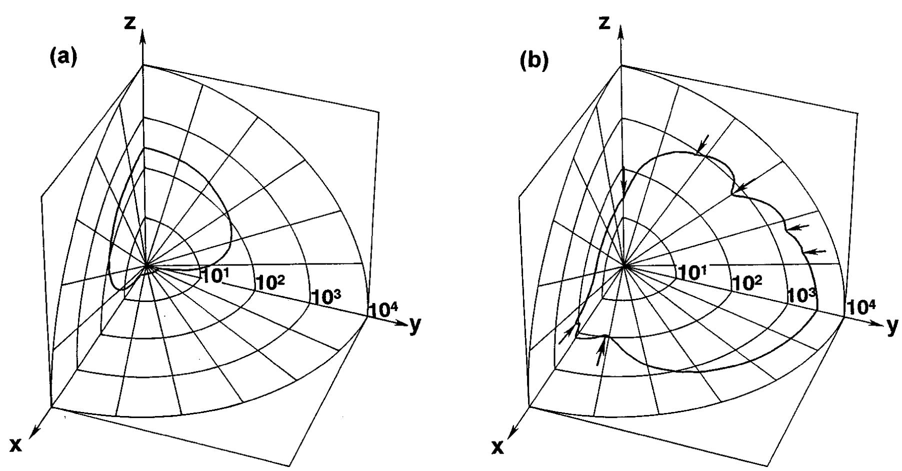


FIG. 1. Calculated angular dependence of the diagonal resistivity elements in the Q1D system. $\pi/(\hbar^2/2t_a a^2 eB) = 20$ is assumed. (a) Normalized interchain resistivity in the conducting layer $\rho_{yy}/(\pi abcB/2e)$. (b) Normalized interlayer resistivity $\rho_{zz}/(\pi abcB/2e)$. Arrows in the *y*-*z*, *x*-*z*, and *x*-*y* plane indicate the Lebed resonance, the Danner-Chaikin oscillation, and the kink structure of the third angular effect, respectively.

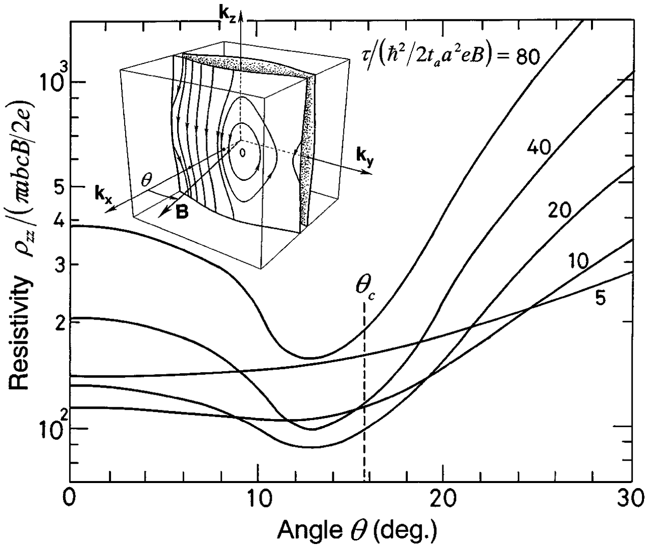


FIG. 2. Dependence of the third angular effect on the parameter $\tau/(\hbar^2/2t_a a^2 eB)$. The field angle is measured by the angle $\theta$ between the 1D axis and the magnetic field direction. Dashed line indicates the critical angle $\theta_c$ calculated by (5). Inset: Semiclassical electron orbits on the sheetlike FS's under the magnetic field.

and finally vanish at a critical angle $\theta_c$. In the case of large anisotropy ($t_a \gg t_b \gg t_c$), $\theta_c$ is given by

$$\tan\theta_c = 2t_b b/\hbar v_F\,. \tag{4}$$

Here $v_F$ is the Fermi velocity. If a quarter of the band (1) is occupied, (4) becomes

$$\tan\theta_c = \sqrt{2}\,\frac{t_b}{t_a}\,\frac{b}{a}\,, \tag{5}$$

since $v_F = \sqrt{2}\,t_a a/\hbar$. Around this critical angle $\theta_c$, some kind of structure is expected to appear on the angular dependence of MR. In fact, the kink structure in Fig. 2 locates just below the critical angle calculated from (5). Strictly to say, the critical angle corresponds not to the kink angle but to the angle below which the MR deviates from the monotonous dependence. The third angular effect physically originates from the vanishing (or appearance) of the closed cyclotron orbits on the FS's.

The above mechanism should work also for the *x-z* plane rotation where the Danner-Chaikin oscillations are observed. In fact, Danner *et al.* found a narrow peak structure in $\rho_{zz}$ at the angle where the field direction is parallel to the 1D axis [4]. They ascribed this peak to the resistance enhancement due to closed orbits and reproduced it by numerical calculation. Blundel and Singleton have also referred to this effect [8]. This effect is completely analogous to the third angular effect in the *x-y* plane rotation. In the third angular effect, the peak structure is much wider and its edge is well defined by the kink structure.

We can utilize the third angular effect to estimate the band anisotropy in the most conducting plane in the Q1D conductors. If the lattice parameters and the band filling are known in advance, we can roughly estimate $t_b/t_a$ from the critical angle obtained from the experiment. We have to notice that it has some ambiguity to determine the critical angle from the kink position observed in the experiment.

The third angular effect discussed above is considered to be the most plausible mechanism for the anomalous angular dependence of MR observed in $(DMET)_2I_3$ [7]. Assuming that the observed kink at $\theta = 15°$ is the critical angle of the third angular effect, we can roughly estimate the anisotropy of the conducting layer of $(DMET)_2I_3$ as 1:0.2. Although the electronic structure of $(DMET)_2I_3$ has not been established yet, this value seems not so bad considering the relatively small conductivity anisotropy (10:1) of this Q1D conductor.

However, the magnetotransport behaviors of $(DMET)_2I_3$ are not necessarily consistent with the present model for the third angular effect. As mentioned before, neither the Lebed resonances nor the Danner-Chaikin oscillations have been observed in $(DMET)_2I_3$. This fact is hardly explained by the present model since the Q1D system showing the third angular effect should also show the other two angular effects. So there are still other possibilities on the origin of the angular effect observed in $(DMET)_2I_3$. In this sense $(DMET)_2I_3$ is not the best material to study the third angular effect itself.

In order to obtain the clearer experimental evidence of the third angular effect, we searched for this phenomenon in another organic Q1D conductor $(TMTSF)_2ClO_4$. In contrast with $(DMET)_2I_3$, $(TMTSF)_2ClO_4$ has a well established electronic structure and shows both of the Lebed resonance and the Danner-Chaikin oscillation. So $(TMTSF)_2ClO_4$ is a more suitable material to study the third angular effect.

We measured the interlayer MR of $(TMTSF)_2ClO_4$ as a function of the magnetic field direction. The single crystal samples were cooled down slowly to reach the "relaxed state." The magnetic field was rotated in the most conducting **a**-**b**$'$ plane. The field direction was measured by the angle $\theta$ between the **a** axis (1D axis) and the field direction. The current direction was parallel to the $\mathbf{c}^*$ axis perpendicular to the conducting plane. The measurements were performed at 1.7 K under the fields up to 12 T. In this experimental condition, the system lies in the normal metallic phase.

Figure 3 shows the angular dependence patterns of the interlayer MR of $(TMTSF)_2ClO_4$ at several field values. At low magnetic fields the MR shows monotonous angular dependence. As the field strength is increased, kink structures around $\theta = \pm 10°$ become visible and sharper. These features are similar to those of Fig. 2. The observation of the kink structure in $(TMTSF)_2ClO_4$ which shows the other two angular effects is very consistent with the numerical prediction that the anisotropic Q1D conductor should show the third angular effect in addition to the Lebed resonance and the Danner-Chaikin oscillation.

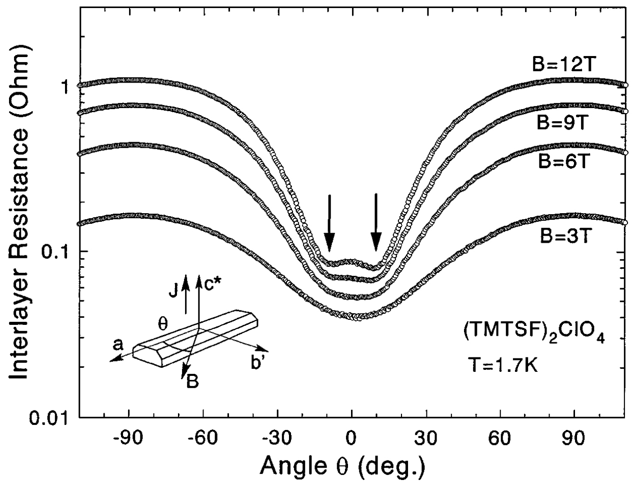


FIG. 3. Angular dependence of the interlayer MR in $(TMTSF)_2ClO_4$ when the magnetic field is rotated in the most conducting plane (**a**-**b**$'$ plane). Arrows indicate the kink structures due to the third angular effect.

Therefore the observed phenomenon is consistently explained by the third angular effect. We consider that the third angular effect has been established as a FS topological effect in the Q1D conductors by the present experiment.

Recently, Yoshino *et al.* independently have observed the precursor of the third angular effect in $(TMTSF)_2ClO_4$ below 7 T. As their magnetic field was too low to see the clear kink structure, they observed the anomaly in the first derivative pattern of the angular dependence of MR [9].

Although the estimation of the anisotropy in $(TMTSF)_2ClO_4$ seems to be a good test for the present picture, the situation is not simple in this compound. In the relaxed state of $(TMTSF)_2ClO_4$, a pair of the original sheetlike FS's are cut into two pairs of sheetlike FS's by the anion ordering superlattice gaps [10]. In this case the third angular effect also occurs, but the anisotropy in the conducting plane cannot be estimated from the critical angle.

In conclusion, we have theoretically and experimentally established the third angular effect of MR as a novel FS topological effect in the Q1D conductors. By the numerical calculation we have shown that the three angular effects, the Lebed resonance, the Danner-Chaikin oscillation, and the third angular effect, appear on the interlayer MR in the anisotropic Q1D conductor . We have experimentally proved this fact using the most studied organic Q1D conductor $(TMTSF)_2ClO_4$. The third angular effect originates from the appearance or vanishing of the closed cyclotron orbits on the sheetlike FS of the Q1D conductors.

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[1] T. Osada, A. Kawasumi, S. Kagoshima, N. Miura, and G. Saito, Phys. Rev. Lett. **66**, 1525 (1991).
[2] M. J. Naughton, O. H. Chung, M. Chaparala, X. Bu, and P. Coppens, Phys. Rev. Lett. **67**, 3712 (1991).
[3] W. Kang, S. T. Hannahs, and P. M. Chaikin, Phys. Rev. Lett. **69**, 2827 (1992).
[4] G. M. Danner, W. Kang, and P. M. Chaikin, Phys. Rev. Lett **72**, 3714 (1994).
[5] K. Maki, Phys. Rev. B **45**, 5111 (1992).
[6] T. Osada, S. Kagoshima, and N. Miura, Phys. Rev. B **46**, 1812 (1992).
[7] H. Yoshino, K. Saito, K. Kikuchi, H. Nishikawa, K. Kobayashi, and I. Ikemoto, J. Phys. Soc. Jpn. **64**, 2307 (1995).
[8] S. J. Blundel and J. Singleton, Phys. Rev. B **53**, 5609 (1996).
[9] H. Yoshino, K. Saito, H. Nishikawa, K. Kikuchi, K. Kobayashi, and I. Ikemoto, Abstract of the Meeting of the Physical Society of Japan (51st Annual Meeting, Part 2), p. 387 (March, 1996).
[10] On the anion ordering in $(TMTSF)_2ClO_4$, see, for example, T. Ishiguro and K. Yamaji, *Organic Superconductors* (Springer-Verlag, Berlin, 1990), and references therein.