Contribution of small closed orbits to magnetoresistance in quasi-two-dimensional conductors

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We find a striking peak structure in the magnetoresistance of the quasi-two-dimensional conductors β-(BEDT-TTF)$_2$I$_3$ and α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ for magnetic fields nearly parallel to the conducting plane. The peak structure can be ascribed to a Fermi-surface topological effect due to the small closed orbits on the side of the warped cylindrical Fermi surface. This effect provides a clue about how to evaluate the interlayer transfer integral. [S0163-1829(98)06503-5]

Organic conductors provide electron systems with low-dimensional Fermi surfaces of quite simple topology, and reveal important aspects overlooked in studies of condensed-matter physics. In quasi-one-dimensional conductors, interesting angular effects of magnetoresistance due to the Fermi surface topology have been discovered.[1–4] In quasi-two-dimensional conductors a similar phenomenon has been found in the interlayer resistance under the magnetic fields rotated in the plane normal to the most conducting plane. It is explained in terms of a kind of commensurability effect between the electron orbits on the Fermi surface under the magnetic fields and the slightly warped cylindrical Fermi surface.[5–7]

Kartsovnik *et al.* also found a peak structure in the magnetoresistance in quasi-two-dimensional conductors when the magnetic field is nearly parallel to the conducting plane.[6,8] It has been proposed that this peak is due to the open orbits on the cylindrical Fermi surface.[9]

In this Brief Report we present similar peak structures found in β_H-(BEDT-TTF)$_2$I$_3$ and α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$. We demonstrate, on the basis of observed results and semiclassical numerical calculations, that these peak structures can be ascribed to the small closed orbits formed on the side of the warped Fermi surface.

We measured the interlayer resistance of β-(BEDT-TTF)$_2$I$_3$ and α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ by the four-probe method. We applied the magnetic fields to the sample in a clamped pressure cell which could be rotated about two axes perpendicular to one other. We made all measurements of β-(BEDT-TTF)$_2$I$_3$ at low temperatures under 5 kbar in the so-called β_H high-T_c state.[10]

Figure 1(a) shows the angular dependence of the magnetoresistance of β_H-(BEDT-TTF)$_2$I$_3$ for a magnetic field rotated in the plane perpendicular to the conducting plane. One finds a series of peaks in the magnetoresistance in the angular range between $\theta=-80°$ and 80°, where θ denotes the angle of the magnetic-field direction measured from the normal to the conducting plane. These peaks are ascribed to Yamaji oscillations, and show the presence of the quasi-two-dimensional Fermi surface,[7] in agreement with the band calculations and Shubnikov–de Haas oscillations.[11,12] The details of the shape of the Fermi surface mapped out will be reported in a separate paper.

Besides this conventional phenomenon, one finds a striking peak structure at $\theta=90°$. These peaks are observed at every azimuthal angle ϕ. This suggests that these peaks have a close relation with the quasi-two-dimensional electron system.

In Fig. 1(b) we show the peak structure measured under various strength of the magnetic field. The peak appears at about 5 T and its height increases with increasing magnetic-field strength. In Fig. 1(b′) we show the comparison of these peaks after normalizing their peak height. It is evident that the peak shape and its angular width between the resistance minima is independent of the magnetic-field strength. This result suggests a change of electronic states caused by some geometrical effect.

We examine the topology of the Fermi surface. As depicted in the inset of Fig. 2(a), the electron orbits generally go around the cylindrical Fermi surface under the fields. However, when the field direction is nearly parallel to the conducting plane, two kinds of peculiar orbits appear on the side of the Fermi surface. One is the open orbit parallel to the cylinder axis of the Fermi surface, and the other is the small closed orbit. The former, proposed by Kartsovnik *et al.*, is expected to increase the magnetoresistance under high magnetic fields, because it causes nonsaturating magnetoresistance. The latter is also considered to enhance the resistance. Under high magnetic fields, these electronic states move in small closed orbits, and their Fermi velocity is averaged to leave only the component parallel to the field direction. The interlayer electrical conduction due to these orbits is reduced as the field becomes parallel to the conducting plane.

To find the dominant mechanism for the observed peak structure, we make a numerical calculation of the magnetoresistance on the basis of the semiclassical theory of electron kinetics. The model is free-electron-like, with the effective mass m^* in the conducting *ab* plane, and tight-binding-like perpendicular to the plane,

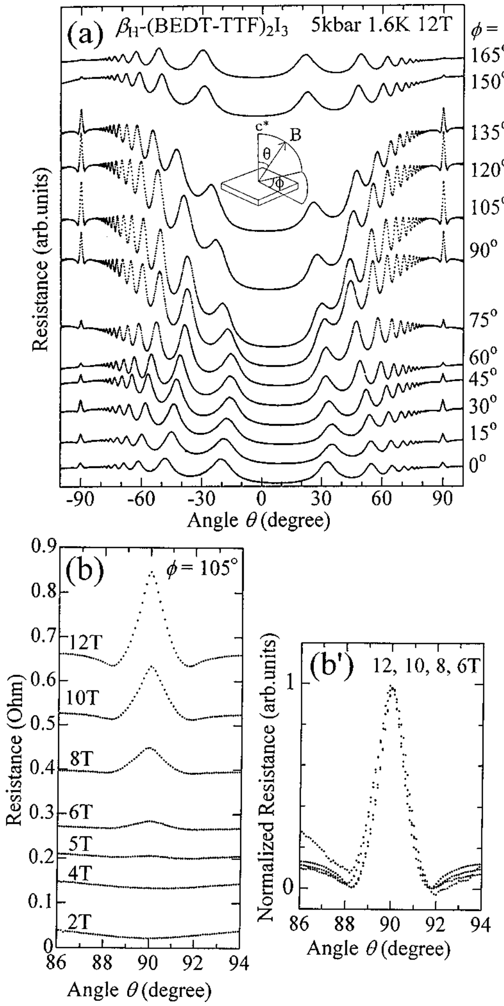

FIG. 1. Angular dependence of the magnetoresistance of β_H-(BEDT-TTF)$_2$I$_3$ for the magnetic field rotated in the plane perpendicular to the conducting plane. (a) Overall features. (b) The peak structure in the magnetoresistance for magnetic fields close to the conducting plane. (b′) The normalized peak shape.

$$E_k = \frac{\hbar^2}{2m^*}(k_x^2 + k_y^2) - 2t_c \cos ck_z. \tag{1}$$

Here k_x, k_y, and k_z denote the a, b, and c components of the wave vector $\mathbf{k}$. t_c is the transfer integral along the c axis. The equation of motion of electrons in the magnetic fields $\mathbf{B}$ is given as usual,

$$\hbar \frac{d\mathbf{k}}{dt} = -e\mathbf{v}\times\mathbf{B}, \quad \mathbf{v} = \frac{1}{\hbar}\frac{dE_k}{d\mathbf{k}}. \tag{2}$$

The conductivity tensor σ_{ij} is calculated using the Boltzmann equation

$$\sigma_{ij} = \frac{2e^2}{V}\sum_k \left[-\frac{df(k)}{dE}\right] v_i(k,0)\,\tau \int_{-\infty}^{0} v_j(k,t)\,\frac{e^{t/\tau}}{\tau}\,dt, \tag{3}$$

where V denotes the volume, $f(k)$ the Fermi distribution function, and v_i the i component of the Fermi velocity, and the relaxation time τ is assumed to be a constant. Here we approximate, for simplicity, the crystalline unit cell as being cubic with the lattice constants $a=b=c=10$ Å. The effective mass is assumed to be the same as the free electron one, $m^* = m_e$, the Fermi wave number $k_F = \pi/2a$, and the ratio of the transfer integrals as $t_a/t_c = 45/1$. Here we introduced t_a to discuss the ratio of the intralayer to interlayer transfer integral. The intralayer transfer integral t_a is defined so that the Fermi velocity becomes equal to that derived from Eq. (1) for the Fermi wave number k_F.

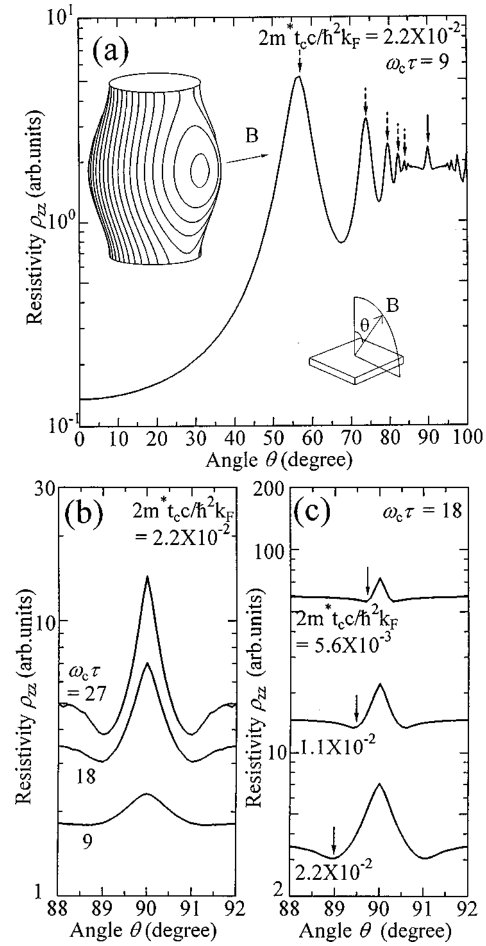

FIG. 2. Angular dependence of the resistance ρ_{zz} calculated on the basis of the semiclassical transport equation. (a) Overall features. The dashed arrows denote the Yamaji oscillations peak, and the solid arrow the peak at $\theta = 90°$. Inset: An example of electron orbits on the quasi-two-dimensional Fermi surface under magnetic fields. (b) Magnetic-field dependence of the peak shape parametrized by the product of the cyclotron frequency and the relaxation time. (c) Transfer integral dependence of the peak shape for various magnitudes of the interlayer transfer integral t_c. The arrows denote the kink structure.

Figure 2(a) shows the calculated results of the angular dependence of the resistance ρ_{zz} derived from the inverse of the conductivity tensor. One finds a sharp kink, and a peak structure near $\theta = 90°$ in addition to the Yamaji oscillations. The calculated results reproduce well the experimental ones shown in Fig. 1(a). Figure 2(b) shows the magnetic-field dependence of the peak shape. The product of the relaxation time τ and the magnetic-field strength represented by the cyclotron frequency $\omega_c = eB/m^*$ is varied, as shown in the figure. The peak is pronounced with increasing field strength, i.e., $\omega_c\tau$ but the angular width of the peak is almost independent of the field strength. These features agree well with experimental results shown in Fig. 1(b). The Yamaji oscillations in Fig. 1(a) suggest the product $\omega_c\tau$ is of the order of 10 at 12 T under the condition of the present study because more than ten oscillation peaks are found there. This $\omega_c\tau$ is evaluated for the cyclotron motion winding the Fermi tube. For cyclotron motions on the side face of the Fermi tube, this product is expected to be reduced by a factor of the order of 10, due to the anisotropy of the transfer integral of the order of 100.

Figure 2(c) shows that the angle of the calculated resistance minimum changes with changing the interlayer transfer integral: the results are calculated for $t_a/t_c=45/1$, 45/0.5, and 45/0.25 and the resistance minima are found near $\theta=89.0°$, 89.5° and 89.7°, respectively. Here we keep the ratio a/c constant.

These features are well explained in terms of a geometrical effect of the closed orbits on the Fermi surface, as shown in the inset of Fig. 2(a). The small closed orbits appear on the side of the cylindrical Fermi surface when the field is perpendicular to a surface element of the Fermi surface. This condition is fulfilled for the field direction close to $\theta=90°$ independent of the field strength. The critical angle θ_c for this is given geometrically as 88.7°, 89.4°, and 89.7° for $t_a/t_c=45/1$, 45/0.5, and 45/0.25, respectively. These critical angles are close to those found in the calculated resistance shown in Fig. 2(c).

Kartsovnik *et al.* ascribed the peak structure to open orbits nearly parallel to the cylinder axis of the Fermi surface.[6,9] It is possible to have these open orbits in a finite angular range of the field direction because of the finite $\omega_c\tau$. If it is the case, however, the angular width of the peak at $\theta=90°$ should be inversely proportional to the magnetic field strength because the open orbits appear for

$$l_k \sin\left(\frac{\pi}{2}-\theta\right) \leqslant 2k_F, \tag{4}$$

where l_k denotes the *mean free path in the k space* which is proportional to $\omega_c\tau$. The magnetic-field dependence of the peak width shown in Fig. 2(b) is much smaller than this estimation. In addition, we think the increase of the ratio t_a/t_c slightly increases the angular width of the peak for the open orbit effect, because the warping of the Fermi surface is reduced. This is again in contradiction with the results in Fig. 2(c).

The calculated resistance and the geometrical consideration above suggest strongly that the remarkable resistance peak at $\theta=90°$ is dominated by small closed orbits formed on the side of the Fermi surface, although the open orbits should also contribute to the peaks.

Let us discuss the contribution of small closed orbits to the magnetoresistance under high magnetic fields. In small closed orbits, the Fermi velocity precesses about the field direction, as shown in the inset of Fig. 2(a). This Fermi velocity is time averaged in the time-integrated part of Eq. (3). As a result, the time-integrated Fermi velocity in these orbits becomes nearly parallel to the field. As the field becomes parallel to the conducting plane, the interlayer component of the time-integrated Fermi velocity is reduced, and the interlayer conductivity decreases. Thus, the peak structure near $\theta=90°$ reflects the dependence of the time-integrated Fermi velocity on the field direction. At the critical angle θ_c, some electronic states have the Fermi velocity perfectly parallel to the field direction. This gives the large current component in the interlayer direction. It is the origin of the resistance minimum.

On the other hand, in the other orbits, the interlayer component of the Fermi velocity oscillates around zero. Thus, it reduces both the interlayer component of the time-integrated part, and also the contribution of these orbits to the interlayer conductivity under the high magnetic fields.

The small closed-orbit effect proposed here allows us to evaluate experimentally the ratio of the intralayer and interlayer transfer integrals, t_a/t_c. Geometrical analysis gives the critical angle θ_c above which the resistance increases rapidly.

$$\frac{\pi}{2}-\theta_c=\frac{2m^*t_c c}{\hbar^2 k_F}\simeq\frac{t_c c}{t_a a}\,\frac{1}{\sin(ak_F)}. \tag{5}$$

Using the parameters $c/a\simeq 2$, $k_F\simeq\sqrt{2\pi}/a$ and $t_c/t_a\simeq 1/140$ for β_{H}-(BEDT-TTF)$_2$I$_3$,[12] we obtain $(\pi/2)-\theta_c\simeq 1.4°$. This value is in rather good agreement with the result in Fig. 1(b), $(\pi/2)-\theta_c\simeq 1.5°$.

In measurements of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ and α-(BEDT-TTF)$_2$KHg(SCN)$_4$ in the normal metallic state under pressure, we found a small resistance peak in the former compound near $\theta=90°$, but not in the latter.[13] This suggests that the interlayer transfer integral of the former is larger than the latter. This is consistent with the absence of the nesting of the one-dimensional Fermi surface in the former because of the enhancement of the corrugation of the Fermi surface. A detailed comparison will be published elsewhere.

Finally the critical magnetic field at which electrons make a complete closed orbit before scattering is

$$B_c\simeq\frac{2\pi\hbar}{ec\tau}\left(\frac{m^*}{2t_c}\right)^{1/2}. \tag{6}$$

When the field strength is larger than this critical magnetic field, the peak and the kink structure can be observed in the magnetoresistance. Here the scattering time τ is averaged over the closed orbits. The onset field will give a clue about how to evaluate the scattering time in a specific region on the Fermi surface.

One should note that similar peak structures have been found in quasi-one-dimensional conductors such as (TMTSF)$_2X$. The Danner oscillations and the third angular effect show the presence of small peaks.[3,4,14] These peaks have the same origin as those in the present study. Similar peak structures in other materials[15] might possess contributions from the small closed-orbit effect presented here. We think this phenomenon is observed not only in the quasi-two-dimensional conductors, but also in any Fermi surface with warping or corrugation.

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