Pulsed magnetic field study of unconventional magnetoresistance of Q1D superconductors 
(TMTSF)$_2$ClO$_4$ and (DMET)$_2$I$_3$

H Yoshino$^1$, Z Bayindir$^2$, J Roy$^3$, B Show$^3$, H-I Ha$^4$, A G Lebed$^5$, M J Naughton$^3$, K Kikuchi$^6$, H Nishikawa$^7$ and K Murata$^1$

1 Graduate School of Science, Osaka City University, Osaka 558-8585, Japan
2 Department of Physics, Queen’s University, Kingston, Ontario, K7L 3N6, Canada
3 Department of Physics, Boston College, Chestnut Hill, MA, 02467, USA
4 Department of Physics, Harvard University, Cambridge, MA 02138, USA
5 Department of Physics, University of Arizona, Tucson, AZ 85721, USA
6 Graduate School of Science and Technology, Tokyo Metropolitan University, Minami-Osawa 192-0397, Japan
7 Department of Chemistry, University of Tsukuba, Tsukuba 305-8571, Japan

E-mail: yoshino@sci.osaka-cu.ac.jp

Abstract. Quasi-one-dimensional (Q1D) organic conductors with simple open Fermi surfaces have provided novel angular dependent magnetoresistance oscillation (AMRO) phenomena for the last two decades. Both semiclassical and quantum mechanical theories well explain these phenomena, but detailed study of the field dependence probably gives a crucial test. Recently it was proposed that a kind of AMRO that is known as “Lee-Naughton oscillations (LNO)” observed for (TMTSF)$_2$ClO$_4$ is a result of dimensional crossover of electronic system from one-to two-dimension on changing the field orientation. We have studied the field dependence of the magnetoresistance of Q1D superconductors (TMTSF)$_2$ClO$_4$ and (DMET)$_2$I$_3$ by using a 46-T pulsed magnetic field system and found the $B^{1/2}$-behavior at minima of LNO.

1. Introduction

Quasi-one-dimensional (Q1D) metals are known to show several kinds of angular dependent magnetoresistance oscillations (AMRO) depending on the rotational axis of the magnetic field at low temperature. The anisotropy of the Q1D electronic system such as (TMTSF)$_2$ClO$_4$ and (DMET)$_2$I$_3$ (TMTSF = tetramethyltetraselenafulvalene, DMET = dimethyl(ethylenedithio)diselenadithiafulvalene) is well described by the tight-binding dispersion relation, $E = -2t_x \cos(k_x x) -3t_y \cos(k_y y) -6t_z \cos(k_z z)$. The ratio of the transfer integrals along the $x$-, $y$- and $z$-axes is typically 300:30:1 and the band-filling of 3/4-electron (or 1/4-hole) results in a pair of open Fermi surfaces.

Some authors [1, 2, 3] have tried to apply quantum mechanics to understand the AMRO phenomena for the last several years, though semiclassical calculations starting from Boltzmann equation also reproduce the structure of each AMRO known as ‘Lebed resonance’, ‘Danner-Kang-Chaikin oscillations, ‘the third angular effect (TAE)’ and ‘Lee-Naughton oscillations (LNO)’. Especially some of the present authors claimed that a series of one-dimensional (1D)
to two-dimensional (2D) crossovers are responsible for LNO [4], which are oscillations of the z-axis resistivity ($\rho_z$) when the magnetic field (typically > 5 T) is rotated around an axis that is slightly ($\lesssim 15^\circ$) tilted from the z*-axis. Indeed saturating behavior of the field dependence of the magnetoresistance $\rho_z(B)$, which was predicted by the theory, has been observed at the minima of LNO in (TMTSF)$_2$ClO$_4$ at 0.1 K and up to 10 T [5]. A semiclassical calculation, however, suggests the possibility of an increase in the slope of $\rho_z(B)$ at much higher magnetic field region. Thus measurements at much higher magnetic fields are required to assess whether the quantum mechanical theory is needed to understand the unconventional transport properties of Q1D systems or not. In this study, we discuss the change in the slope of $\rho_z(B)$ of (TMTSF)$_2$ClO$_4$ and (DMET)$_2$I$_3$ in the LNO angle region up to 40 T for the first time.

2. Experimental

Single crystals of (TMTSF)$_2$ClO$_4$ and (DMET)$_2$I$_3$ were obtained by usual electrochemical oxidation method. Dimension of the sample is $0.98 \times 0.16 \times 0.14$ mm$^3$ for (TMTSF)$_2$ClO$_4$ (#0402) and $0.82 \times 0.20 \times 0.06$ mm$^3$ for (DMET)$_2$I$_3$ (#0502), respectively. It should be noted that the first (x), second (y) and third (z) conducting axes are a, b and c in (TMTSF)$_2$ClO$_4$ and b, a and c in (DMET)$_2$I$_3$. Two pair of annealed gold wires (12.7 $\mu$m in diameter) were attached by using carbon paste on the opposite crystal surfaces corresponding to the xy plane to measure the z*-axis electrical resistivity by the four-probe method. The relaxed (R-) state of (TMTSF)$_2$ClO$_4$, where the anion-ordering transition at 24 K and superconductivity below 1.2 K are observed, was achieved by cooling the sample of (TMTSF)$_2$ClO$_4$ with the cooling rate of less than 5 K/h from 35 to 15 K.

Pulsed magnetic field above 40 T was generated by a commercial system (PulseLab, Oxford Instruments). Typical duration time is in the order of 20 ms. High frequency measurement was carried out by using a combination of a lock-in amplifier (7280, Perkin-Elmer) and a preamplifier (SR560, Stanford Research) for (TMTSF)$_2$ClO$_4$ and another combination of a lock-in amplifier (SR844, Stanford Research), a pulse/function generator (HP8116A, Hewlett-Packard) and a preamplifier (SR560) for (DMET)$_2$I$_3$.

The absolute value of the magnetic field was determined by measuring the induced voltage of a pick-up coil in the vicinity of the samples. The samples were set on a rotating platform of the cryostat insert to change its relative direction to the vertical magnetic field. This is called $\phi$-rotation in this paper. Two series of $\phi$-rotations were performed at two ascending vertical angles $\theta \sim 0^\circ$ and $\sim 5^\circ$ for (TMTSF)$_2$ClO$_4$ by changing the orientation of the crystal at room temperature. Definitions of $\theta$ and $\phi$ relative to the lattice vectors are shwon in figure 1. Magnetic field dependence of the magnetoresistance was measured under the pulsed-field on changing $\phi$ with step of $\sim 1^\circ$ at 4.2 K. Angular dependence was extracted from a series of field dependence.

3. Results and discussion

Figure 2 shows the $\phi$ dependence of the normalized magnetoresistance at $\theta \sim 0^\circ$. High quality data obtained by the pulsed-field enabled us to extract fine structure of the angular dependence.
One can see a broad hump develops between $\phi = -12^\circ$ and $+12^\circ$ on increasing magnitude of the magnetic field. This is the TAE usually observed for Q1D metals when the magnetic field is rotated within the $xy$ plane. Since $\theta$ is almost zero, weak LNO are recognized between $\phi = 5^\circ$ and $12^\circ$ above 25 T. The amplitude of the LNO became much more distinct when $\theta$ was changed from $0^\circ$ to $5^\circ$ as in figure 3.

The sharp minimum at $\phi = 0^\circ$ and maximum at $\phi = 6^\circ$ in figure 3 should correspond to qualitatively different field dependence of the magnetoresistance if dimensional crossover is expected. This is compared in figure 4. The magnetoresistance at the minimum seems to

Figure 2. Angular dependence of normalized magnetoresistance of (TMTSF)$_2$ClO$_4$ in the R-state at 4.2 K and $\theta \sim 0^\circ$ measured by pulsed magnetic field.

Figure 3. Angular dependence of normalized magnetoresistance of (TMTSF)$_2$ClO$_4$ in the R-state at 4.2 K and $\theta \sim 5^\circ$ measured by pulsed magnetic field.

Figure 4. Field dependence of normalized magnetoresistance of (TMTSF)$_2$ClO$_4$ in the R-state at 4.2 K. Data at $\phi = 0^\circ$ and $6^\circ$ correspond to the minimum and maximum of the angular dependence in figure 3.

Figure 5. Normalized magnetoresistance of (TMTSF)$_2$ClO$_4$ in the R-state at 4.2 K plotted against $B^{1/2}$. The data are the same as that at $\phi = 0^\circ$ in figure 4. The solid line is a guide to the eye.
saturate, while the slope of the magnetoresistance at the maximum continues to increase even at 40 T. So apparently this is consistent with the dimensional crossover picture. The field dependence of the magnetoresistance at $\phi \sim 0^\circ$ is, however, found to be not saturating but proportional to $B^{1/2}$ as in figure 5. The magnetoresistance is plotted against square root of the magnetic field. The data seems to be well fitted by linear line above 9 T. This is also the case at the minimum ($\phi = 11^\circ$) in figure 2.

Since (TMTSF)$_2$ClO$_4$ has two pairs of Q1D Fermi surfaces in the R-state, there might occur mixing of different types of field dependences. Therefore the same kind of experiment was carried out for another Q1D metal (DMET)$_2$I$_3$ that has only one pair of open Fermi surfaces. The angular dependence is shown in figure 6. The field dependence at one of minima ($\phi = 2^\circ$) in figure 6 is plotted against $B^{1/2}$ in figure 7. Again very good linearity is seen above 4 T. Thus we can probably conclude that $B^{1/2}$-behavior is common at the field orientation corresponding to TAE and LNO minima. This type of field dependence has been investigated theoretically for $\theta = 0^\circ$, namely, at the minima of TAE [6, 7]. We found experimentally that the $B^{1/2}$-behavior is also observed at the LNO minima in this study.

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