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1992 J. Phys.: Condens. Matter 4 525

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## A new magneto-optic effect of quasi-two-dimensional conductors

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Received 31 May 1991, in final form 3 September 1991

**Abstract.** The formation of magnetic-field-induced spin-density-wave phases with quantized wavevectors in a quasi-two-dimensional organic conductor results in the quantization of the Hall conductance. In this paper we study the rotation of a polarized electromagnetic wave incident perpendicular to the molecular layer at a moderate magnetic field. Some interesting features are shown which should be useful in the investigation of Bechgaard salts.

Two kinds of quantized Hall effect have been discovered. One is the integer and fractional quantized Hall effect in high-mobility two-dimensional electronic system, formed in metal–oxide–semiconductor field effect transistors or semiconductor heterostructures [1, 2]. This kind of quantized Hall effect arises from the localization of electrons in two-dimensions and the presence of gaps in the quasi-particle excitation spectrum [3]. Whenever the Fermi level lies in a gap of extended states, the longitudinal conductivity  $\sigma_{xx}$  vanishes; concomitantly, the Hall conductivity  $\sigma_{xy}$  is equal to  $fe^2/h$ , with  $f$  being an integer or a rational fraction. Another kind of quantum Hall effect is discovered in the quasi-two-dimensional organic conductors  $(\text{TMTSF})_2\text{X}$ , where TMTSF is tetramethyltetraselenafulvalene and  $\text{X} \equiv \text{PF}_6, \text{ClO}_4, \text{ReO}_4$ , etc [4–11].

In zero field the Bechgaard salts  $(\text{TMTSF})_2\text{X}$  are quasi-two-dimensional open-orbit metals. The application of a magnetic field reduces the dimensionality by one and the resulting one-dimensional metal is unstable against the formation of a spin-density wave of wavevector  $q = 2k_F$ , where  $k_F$  is the wavevector on Fermi surface [12, 13]. The reciprocal of the magnetic length gives another periodicity which competes with  $q$  and produces a series of magnetic-field-dependent Landau bands and gaps in the vicinity of the Fermi energy  $E_F$ . The system lowers its energy by adjusting  $q$  so that  $E_F$  is in the largest gap of a particular field. At low fields there are many filled Landau bands. Increasing the field causes the gap structure to change,  $q$  jumps, and  $E_F$  lies in the next gap with one fewer Landau band filled. The cascade of transitions continues until no filled Landau bands exist and  $E_F$  is in the one large symmetric gap which remains. From the initial transition to the last,  $E_F$  always remains in the gap between Landau

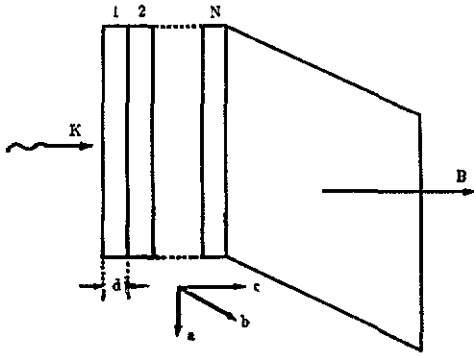


Figure 1. An organic conductor is considered as consisting of  $N$  two-dimensional electron systems with separation  $d$ . The magnetic field is applied perpendicular to the  $a$ - $b$  plane. We take the  $z$  axis along the direction of the  $c$  axis. The incident radiation is propagating along the magnetic field.

levels, producing a quantized Hall conductance, until the final semiconducting state is reached at high fields.

In a previous paper [14], we studied the Faraday rotation of a quasi-two-dimensional electron system in the quantized Hall regime and showed that in the high-frequency regime the Faraday rotation is quantized to the fine structure constant  $\alpha = e^2/\hbar c$ , whereas in the low-frequency limit one circular polarization of radiation is transparent while another is absorbed. In the present paper, we are interested in Faraday rotation in the quasi-two-dimensional organic conductor  $(\text{TMTSF})_2\text{X}$  in the magnetic-field-induced spin-density wave regime which shows quantization of the Hall conductance. The frequency dependence is calculated using standard analysis as in conventional semiconductor systems. We show that a finite value of the longitudinal conductivity modifies the picture of quantization in the high-frequency regime although the appearance of plateaux in Faraday rotation is unaffected.

We consider a simple model of an orthorhombic anisotropic quasi-two-dimensional  $(\text{TMTSF})_2\text{X}$  organic conductor. The most conducting plane is within the  $a$ - $b$  plane. Perfect nesting along the  $c$  direction makes the problem effectively two dimensional. Consequently we view the conductor as a stack of  $N$  two-dimensional electron systems with separation  $d$ , where  $d$  is the lattice parameter in the  $c$  direction. The magnetic field is applied parallel to the  $c$  direction (figure 1). Faraday rotation describes the rotation of the polarization of radiation passing through the conductor along the direction of the applied magnetic field [15]. Physically it arises from the differences in propagation of the two senses of circular polarization into which the plane-polarized beam may be resolved. The Faraday rotation  $\theta$  of  $N$  molecular layers of thickness  $d$  is defined as

$$\theta = (N\omega d/2c)(\eta_- - \eta_+) \quad (1)$$

where  $\eta_+$  and  $\eta_-$  are the indices of refraction of right and left circularly polarized radiation of frequency  $\omega$ . Expressions for  $\eta_{\pm}$  can be easily obtained using Maxwell's equations

$$\nabla \times \mathbf{E} = -\dot{\mathbf{B}}/c \quad \nabla \times \mathbf{H} = 4\pi\mathbf{J}/c + \dot{\mathbf{D}}/c \quad (2)$$

which combined with the constitutive equations

$$\mathbf{J} = \boldsymbol{\sigma} \cdot \mathbf{E} \quad \mathbf{B} = \mu\mathbf{H} \quad \mathbf{D} = \epsilon\mathbf{E} \quad (3)$$

give

$$\nabla(\nabla \times \mathbf{E}) - \nabla^2\mathbf{E} = -(4\pi\mu/c^2)\boldsymbol{\sigma} \cdot \dot{\mathbf{E}} - (\mu\epsilon/c^2)\ddot{\mathbf{E}}. \quad (4)$$

For the Faraday effect, one takes the propagation along the applied magnetic field. Assuming a plane-wave solution of the form

$$\mathbf{E} = \mathbf{E}_0 \exp[i(\omega t - \mathbf{K} \cdot \mathbf{z})] \quad (5)$$

leads to

$$\mathbf{K}^2 \mathbf{E}_0 = \mu \varepsilon (\omega/c)^2 [1 - (4\pi i/\omega \varepsilon) \boldsymbol{\sigma}] \mathbf{E}_0 \quad (6)$$

where  $\mathbf{1}$  is a unit tensor. On the assumption of a circular polarized wave  $\mathbf{E}_0 = E_{0x} \pm i E_{0y}$ , then

$$K_{\pm}^2 = \mu \varepsilon (\omega/c)^2 [1 - (4\pi i/\omega \mu) \sigma_{\pm}^{(3D)}] \quad (7)$$

where  $\sigma_{\pm}^{(3D)} = \frac{1}{2}(\sigma_{xx}^{(3D)} + \sigma_{yy}^{(3D)}) \pm \frac{1}{2}\sqrt{(\sigma_{xx}^{(3D)} - \sigma_{yy}^{(3D)})^2 + 4\sigma_{xy}^{(3D)}\sigma_{yx}^{(3D)}}$ . The complex refractive index  $\eta - i\kappa$  is obtained from

$$(\eta_{\pm} - i\kappa_{\pm})^2 = \mu \varepsilon [1 - (4\pi i/\omega \varepsilon) \sigma_{\pm}^{(3D)}]. \quad (8)$$

When the applied magnetic field is low, the  $(\text{TMTSF})_2\text{X}$  organic conductor is in the metallic phase and shows no quantum Hall behaviour. Above a threshold magnetic field, cooperative effects of electron interactions related to the Fermi-surface anisotropy modify the picture. Diamagnetic effects pin the Fermi energy in a Landau gap and electron-hole pairing leaves only small pockets of itinerant states. These effects lead to a cascade of field-induced first-order transitions and quantized Hall conductance in each molecular layer [12, 13]. Within a molecular layer, the two-dimensional conductivity tensor takes the form

$$\boldsymbol{\sigma}^{(2D)} = \begin{bmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yz} & \sigma_{yy} \end{bmatrix} \quad (9)$$

with  $\sigma_{xy} = -\sigma_{yx} = fe^2/h$ . Here the Landau filling factor  $f$  is an integer or even a rational fraction [10, 11]. It should be mentioned that the diagonal conductivity  $\sigma_{xx}$  or  $\sigma_{yy}$  in  $(\text{TMTSF})_2\text{X}$  organic conductors is finite in contrast with the vanishing of the diagonal conductivity in the usual quantum Hall effect observed in the 2D electron system in a MOSFET or a heterostructure. The three-dimensional conductivity is related to the two-dimensional conductivity and the separation  $d$  between molecular layer by the following expression:

$$\boldsymbol{\sigma}^{(3D)} = \boldsymbol{\sigma}^{(2D)}/d. \quad (10)$$

The electromagnetic radiation is incident along the direction of the magnetic field pictorially shown in figure 1. We assume that the temperature and the intensity of radiation are so low that the transport characteristics of the  $(\text{TMTSF})_2\text{X}$  organic conductor is unaffected. To the best of our knowledge, there exists as yet no theory on the interaction of the field induced spin-density wave ground states, believed to be responsible for the quantum Hall behaviour, with electromagnetic radiation. Therefore the results presented here are somewhat speculative. However, optical investigations of the integer and fractional quantum Hall effect in an ultrahigh-mobility two-dimensional electronic system have demonstrated that temperature rise and the presence of photoexcited holes modify the depth of the  $\sigma_{xx}$  minima but do not significantly affect the low-temperature transport characteristics of the system [16-21].

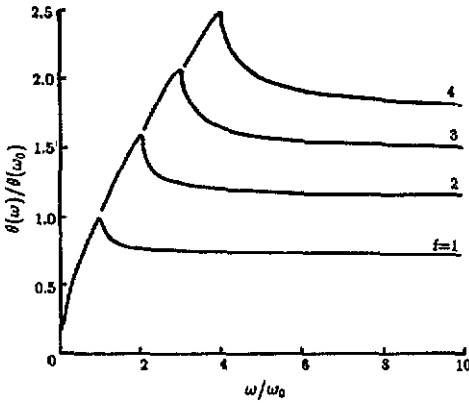


Figure 2. Slopes of the normalized Faraday rotation  $\theta(\omega)/\theta(\omega_0)$  for various Landau indices as a function of the normalized frequency  $\omega/\omega_0$ . Here  $f$  is the Landau index,  $\omega_0 = 4\pi e^2/h\epsilon d$  is the characteristic frequency, and the parameter  $\lambda$  is set at 100.

Now we calculate the Faraday rotation  $\theta$  in the magnetic-field-induced spin-density-wave phases. We introduce the following notation:  $\bar{\sigma}_{xx} = \frac{1}{2}(\sigma_{xx} + \sigma_{yy})$  and  $\bar{\sigma}_{xy} = \frac{1}{2}\sqrt{4\sigma_{xy}^2 - (\sigma_{xx} - \sigma_{yy})^2}$ . Combining equations (8)–(10) we have

$$(\eta_{\pm} - i\kappa_{\pm})^2 = \mu\epsilon[(1 \pm 4\pi\bar{\sigma}_{xy}/d\omega\epsilon) - 4\pi i\bar{\sigma}_{xx}/d\omega\epsilon] \quad (11)$$

from which  $\eta_{\pm}$  and  $\kappa_{\pm}$  are obtained:

$$\eta_{\pm} = (\eta/\sqrt{2})[\sqrt{(1 \pm 4\pi\bar{\sigma}_{xy}/\omega\epsilon d)^2 + (4\pi\bar{\sigma}_{xx}/\omega\epsilon d)^2} + (1 \pm 4\pi\bar{\sigma}_{xy}/\omega\epsilon d)]^{1/2} \quad (12a)$$

$$\kappa_{\pm} = (\eta/\sqrt{2})[\sqrt{(1 \pm 4\pi\bar{\sigma}_{xy}/\omega\epsilon d)^2 + (4\pi\bar{\sigma}_{xx}/\omega\epsilon d)^2} - (1 \pm 4\pi\bar{\sigma}_{xy}/\omega\epsilon d)]^{1/2} \quad (12b)$$

where  $\eta = \sqrt{\mu\epsilon}$  is the refractive index. The Faraday rotation can be calculated with the use of the definition in equation (1).

Specifically if we assume  $\sigma_{xx} = \sigma_{yy}$  so that  $\bar{\sigma}_{xx} = \sigma_{xx}$ ,  $\bar{\sigma}_{xy} = fe^2/h$  and introduce the parameter  $\lambda = (e^2/h)/\sigma_{xx}$  within each molecular layer and a characteristic frequency  $\omega_0 = 4\pi e^2/h\epsilon d$ , the Faraday rotation is then determined:

$$\theta = (\eta N\omega d/2^{3/2}c)\{[\sqrt{(1 - f\omega_0/\omega)^2 + (\omega_0/\lambda\omega)^2} + (1 - f\omega_0/\omega)]^{1/2} - [\sqrt{(1 + f\omega_0/\omega)^2 + (\omega_0/\lambda\omega)^2} + (1 + f\omega_0/\omega)]^{1/2}\} \quad (13)$$

where  $Nd$  is the thickness of the crystal and  $c$  the velocity of light.

Recent experiments on a single crystal of the organic conductor  $(\text{TMTSF})_2\text{PF}_6$  have shown that there are well defined peaks of longitudinal resistance at the fields where the Hall voltage jumps to the next plateau [8, 9]. Although the temperature dependences of the longitudinal resistance and the Hall resistance are different from those found for the conventional quantum Hall effect, the longitudinal resistance in the region between peaks is constant and corresponds to a resistance which is 150–200 times lower than  $h/e^2$  per molecular layer. If we take the  $c$  lattice parameter [8, 20] to be 13.3 Å for  $(\text{TMTSF})_2\text{PF}_6$  and invoke the simple model equation (13),  $\omega_0 = 4\pi e^2/h\epsilon d$  is of the order of  $10^{15} \text{ s}^{-1}$ , which is approximately within the region of visible light. Consequently it may be concluded from equation (13) that the Faraday rotation at constant frequency will show plateaux as the magnetic field is changed provided that the Fermi energy is pinned in a gap of spin-density wave phase, and the Hall conductivity takes the form  $\sigma_{xy} = fe^2/h$  with  $f$  an integer. The frequency dependence for several Landau indices is plotted in figure 2. Here the parameter  $\lambda$  is taken to be 100 as obtained from experimentation. For small  $\omega/\omega_0$  the Faraday rotation  $\theta$  tends to be very small and differences between

neighbouring Landau indices are indistinguishable. After an initial increase with increasing frequency a peak is reached for a given Landau index, which provides a determination of the characteristic frequency  $\omega_0 = 4\pi e^2/h\epsilon d$ . For large  $\omega/\omega_0$ , the Faraday rotation  $\theta$  approaches a constant value and shows little frequency dependence. When  $\lambda$  tends to be very large corresponding to the vanishing of  $\sigma_{xx}$ ,  $|\theta|$  reduces to the value  $(2\pi Nf/\epsilon)\alpha$  with  $\alpha = e^2/hc$ , the fine-structure constant. This is the expected behaviour for the conventional quantum Hall effect of a MOSFET or a heterostructure [14].

In conclusion we have calculated the behaviour of polarized radiation passing through a  $(\text{TMTSF})_2\text{X}$  single crystal in the spin-density-wave phases. Although the general behaviour of polarized radiation in such a system is unknown, quantization of Hall conductance results in plateaux in the Faraday rotation as the applied magnetic field varies. The finite longitudinal conductivity causes a deviation from perfect quantization of the Faraday rotation in the high-frequency regime. These results are applicable to the study of the  $(\text{TMTSF})_2\text{X}$  organic conductor.

### Acknowledgment

This work was supported by the Chinese Science Foundation.

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