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1997 J. Phys.: Condens. Matter 9 L47

(http://iopscience.iop.org/0953-8984/9/6/001)

J. Phys.: Condens. Matter **9** (1997) L47–L54. Printed in the UK PII: S0953-8984(97)79535-3

LETTER TO THE EDITOR

The quantum Hall effect in organic metals with both quasi-two-dimensional and quasi-one-dimensional Fermi-surface components

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Received 13 November 1996

Abstract. The quantum Hall effect (QHE) is shown to be a natural property in high magnetic fields of any system possessing both quasi-two-dimensional and quasi-one-dimensional Fermi-surface components. Such Fermi surfaces are known to occur in some organic charge-transfer salts (e.g. α-(BEDT-TTF)$_2$MHg(SCN)$_4$ with $M = K, Tl, NH_4$). Whilst the QHE in two-dimensional semiconductor systems such as heterostructures and MOSFETs relies on the existence of localized states at the edges of the Landau levels, in organic metals the quasi-one-dimensional portion of the Fermi surface is shown to provide the necessary reservoir for pinning the chemical potential between the completely filled and empty Landau levels of the quasi-two-dimensional Fermi-surface section.

The quantum Hall effect has usually been regarded as a property of two-dimensional semiconductor systems such as GaAs–(Ga, Al)As heterostructures or Si MOSFETs [1–3]. However, recent magnetization measurements on the charge-transfer salts α-(BEDT-TTF)$_2$MHg(SCN)$_4$ ($M = K, Tl$) [4, 5] have revealed sharp features apparently due to persistent eddy currents; it was suggested [4] that these were associated with the deep minima in the resistivity component ρ_{xx} which accompany the quantum Hall-effect plateaux [1, 2] in ρ_{xy}. This assertion, if correct, would mark the first observation of the conventional quantum Hall effect in a bulk material [3]. In this letter we report calculations which show that the quantum Hall effect should indeed be a natural property of materials such as α-(BEDT-TTF)$_2$MHg(SCN)$_4$ ($M = K, Tl$).

There are two prerequisites for the observation of the quantum Hall effect. Firstly, the energy separation of the Landau levels should be greater than their width; it is this requirement which rules out the quantum Hall effect in conventional three-dimensional metallic systems because the bandwidth W in the direction in which the field is applied is invariably much greater than the Landau-level separation $\hbar\omega_c$. Secondly, the chemical potential μ should be situated between completely filled and empty Landau levels over an extended region of magnetic field B to give rise to plateaux in ρ_{xy}; this is realized in two-dimensional semiconductor systems because of the localized nature of the states in the tails of the Landau levels [2].

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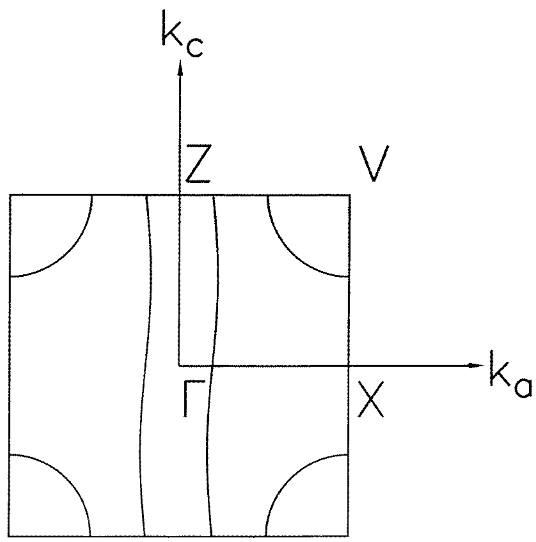

Figure 1. The calculated Fermi surface for the α-phase BEDT-TTF salts, consisting of both quasi-one-dimensional and quasi-two-dimensional sections (after reference [8]; see also reference [9]).

The α-(BEDT-TTF)$_2$MHg(XCN)$_4$ (M = K, Tl or NH$_4$ and X = S or Se) charge-transfer salts seem to fulfil the first prerequisite admirably [6, 7], often possessing quasiparticle lifetimes τ and bandwidths in the direction perpendicular to the highly conducting planes such that $\omega_c\tau \sim 3$–10 and $\hbar\omega_c \gg W$ at $B \sim 30$ T. However, the situation is complicated in the α-phase salts by the presence of a quasi-one-dimensional component of the Fermi surface [8, 9] in addition to the closed quasi-two-dimensional cylinder which gives rise to the quantized orbital motion (see figure 1). The open Fermi-surface sheets cannot undergo Landau quantization in a magnetic field; hence their contribution to the density of states (DOS) is a continuous function of energy E. Such quasi-one-dimensional states do not occur in semiconductor heterostructures or MOSFETs [2] and so their possible involvement in the quantum Hall effect has not thus far been considered.

Recent numerical calculations [6] have successfully simulated high-field de Haas–van Alphen [12] and Shubnikov–de Haas [13] oscillations in the α-phase BEDT-TTF salts; the DOS used in the calculations contained contributions from both quasi-one-dimensional (constant DOS) and quasi-two-dimensional (Lorentzian Landau-level DOS) Fermi-surface sections. It was found at high magnetic fields that μ remained within the quasi-one-dimensional component of the DOS (i.e. *between* Landau levels) over relatively extended regions of B (see especially figure 1 of reference [6]). Thus it does appear that both prerequisites for the quantum Hall effect can be fulfilled in the α-phase BEDT-TTF salts. Nevertheless, as the quantum Hall effect is usually observed in measurements of the resistivity, it might at first be thought that contributions to the electrical conductivity due to the quasi-one-dimensional parts of the Fermi surface would in some way act to 'short out' or obscure effects due to the quantized conductivity of the two-dimensional cylinders; as will be demonstrated below, this turns out not to be the case.

We shall first consider the effect of ideally one-dimensional Fermi-surface sheets; in this case, the corresponding conductivity components $\sigma_{xy,1D}$ and $\sigma_{yy,1D}$ of the one-dimensional states are zero, whilst the $\sigma_{xx,1\mathrm{D}}$ contribution remains finite. In the situation where μ is located directly between adjacent Landau levels, the contributions from the quasi-two-dimensional Fermi-surface sections will be $\sigma_{xx,2\mathrm{D}} = \sigma_{yy,2\mathrm{D}} = 0$ and $\sigma_{xy,2\mathrm{D}} = eN_{2\mathrm{D}}/B$ (here $N_{2\mathrm{D}}$ is the areal density of the quasi-two-dimensional carriers), and it is straightforward to show that

$$\begin{vmatrix} \rho_{xx} & \rho_{xy} \\ \rho_{yx} & \rho_{yy} \end{vmatrix} = \begin{vmatrix} 0 & R_{\mathrm{H}} \\ -R_{\mathrm{H}} & R_{\mathrm{H}}^2\sigma_{xx,1\mathrm{D}} \end{vmatrix} \tag{1}$$

with $eN_{2\mathrm{D}}/B = 1/R_{\mathrm{H}}$. Whilst the diagonal elements are clearly anisotropic, the off-diagonal Hall components have the conventional form expected for a quasi-two-dimensional Fermi-surface pocket [2]. We now consider an extended region of B over which μ resides in the quasi-one-dimensional DOS (see figure 1 of reference [6]); this has the effect of keeping the Landau levels below (above) μ completely filled (empty). Throughout this interval, the Landau levels have degeneracy $d = 1/(2\pi\lambda^2)$, where $\lambda = (\hbar/(eB))^{1/2}$; hence from summing over all occupied Landau levels, the total number of quasi-two-dimensional states is $N_{2\mathrm{D}} = ieB/h$, i being the index of the uppermost filled Landau level. Substituting this into the off-diagonal elements of equation (1) produces

$$\rho_{xy} = -\rho_{yx} = \frac{h}{ie^2} \tag{2}$$

i.e. the quantum Hall effect.

The finite warping of the quasi-one-dimensional sections of Fermi surface in the α-phase BEDT-TTF salts implies a departure from this ideal situation. However, it will be shown below that the quantum Hall effect is still tenable provided that the conductivity of the quasi-one-dimensional Fermi-surface section is sufficiently anisotropic. Quasi-one-dimensional sections of Fermi surface are habitually modelled by the equation [14]

$$E = hv_{\mathrm{F}}(k_x - k_{\mathrm{F}}) - 2t_{\mathrm{c}}\cos(ck_y) \tag{3}$$

where v_{F} is the Fermi velocity and the warping in the direction perpendicular to the conducting planes and higher-harmonic contributions to the warping in the planes are ignored. The wavevectors k_x, k_y and k_z are associated with the lattice parameters a, c and b respectively [8], and the magnetic field is chosen to be in the z-direction; t_{c} is the transfer integral which describes the warping of the quasi-one-dimensional Fermi-surface component within the conducting planes. The classical conductivity tensor for a quasi-one-dimensional Fermi surface is known to take the form [14, 15]

$$\begin{vmatrix} \sigma_{xx,1\mathrm{D}} & \sigma_{xy,1\mathrm{D}} \\ \sigma_{yx,1\mathrm{D}} & \sigma_{yy,1\mathrm{D}} \end{vmatrix} = \sigma_{1\mathrm{D},0} \begin{vmatrix} 1 & -\gamma_{\mathrm{an}}/(\omega_{1\mathrm{D}}\tau) \\ \gamma_{\mathrm{an}}/(\omega_{1\mathrm{D}}\tau) & \gamma_{\mathrm{an}}/(1+\omega_{1\mathrm{D}}^2\tau^2) \end{vmatrix} \tag{4}$$

where

$$\gamma_{\mathrm{an}} = \frac{2c^2t_{\mathrm{c}}^2}{\hbar^2 v_{\mathrm{F}}^2} \tag{5}$$

is the zero-field conductivity anisotropy ratio [14], $\sigma_{1\mathrm{D},0} = e^2N_{1\mathrm{D}}\tau/m_{1\mathrm{D}}^*$ is the zero-field Drude-like conductivity, and $\omega_{1\mathrm{D}} = eB/m_{1\mathrm{D}}^*$ (with $m_{1\mathrm{D}}^* = \hbar/(cv_{\mathrm{F}})$) is the frequency at which the quasiparticles sweep across the quasi-one-dimensional Fermi surface in a magnetic field. Consequently, the extent to which the warping affects the quantum Hall effect depends on the magnitude of γ_{an}.

From equation (3) it follows that t_{c} is related to the width δk_a (figure 1) of the warping of the quasi-one-dimensional Fermi surface by the relationship $\delta k_a = 4t_{\mathrm{c}}/(\hbar v_{\mathrm{F}})$. Combining this with equation (5), we obtain $\gamma_{\mathrm{an}} = \pi^2(\delta k_a)^2/(2k_{\mathrm{c}}^2)$, which is independent of the sizes of t_{c} and v_{F}. This result is rather convenient, since it makes the anisotropy condition independent of band renormalization effects. For example, analysis of the band-structure calculations [8] represented in figure 1 yields $t_c \sim 8$ meV and $v_F \sim 2.5 \times 10^5$ m s^{-1}, values which are somewhat higher than those which might be expected from simple fermiological considerations [17]; nevertheless, the calculated Fermi-surface shape [8, 9] can still be used to give $\gamma_{an} = 10^{-2}$–10^{-3}.

In principle, it would be possible to infer a value of γ_{an} using angle-dependent magnetoresistance oscillation (AMRO) measurements. However, the warping of the quasi-one-dimensional sheets in the α-phase BEDT-TTF salts is thought to be too weak to produce AMROs [18], supporting the very high degree of anisotropy that we have deduced using the calculated Fermi surface.

In order to use this estimate of the anisotropy to assess the feasibility of the quantum Hall effect in a system consisting of both quasi-two-dimensional and quasi-one-dimensional carriers we have performed numerical calculations of the magnetoresistance at high magnetic fields ~30–120 T. As a first approximation we assume that the conductivity of the quasi-one-dimensional Fermi surface behaves in the classical manner described by equation (4). In contrast, the quasi-two-dimensional carriers will depart significantly from the classical situation in the limit $\omega_c\tau \sim 3$–10, valid for such high fields [6, 13]. Indeed, a number of different measurements on BEDT-TTF salts [6] have already established that the Lifshitz–Kosevich (LK) formalism used to describe the quantum oscillatory behaviour of properties such as magnetization and resistivity at low magnetic fields ~10 T is no longer appropriate at the high fields used in e.g. references [4] and [13]. For practical reasons, the high-field magnetoresistance experiments which have demonstrated the failure of the LK approach [6, 13] have so far been confined to measurements of the resistivity component ρ_{zz} (i.e. that measured in the longitudinal z-direction); no direct measurements of the transverse resistivity components ρ_{xx} and ρ_{xy} have thus far been made at high magnetic fields. However, in view of the failure of the LK formalism to describe the high-field oscillations in ρ_{zz} [6, 13] it is expected that the oscillations in ρ_{xx} will also depart from LK-like behaviour at high fields. We therefore calculate ρ_{xx} and ρ_{xy} in the high-field limit numerically, using an approach somewhat analogous to that described for ρ_{zz} in reference [6].

Following Ando and Uemura [19] (an approach based on the Kubo formalism), the diagonal components of the conductivity due to the quasi-two-dimensional Fermi-surface pockets in a magnetic field are given by

$$\sigma_{xx,2D} = \sigma_{yy,2D} = \frac{e^2}{\pi^2\hbar}\int\left(-\frac{\partial f}{\partial E}\right)\sum_i\left(i-\frac{1}{2}\right)\frac{(\mathrm{Im}\,G_i)^2}{(\mathrm{Re}\,G_i)^2+(\mathrm{Im}\,G_i)^2}\,\mathrm{d}E \tag{6}$$

Here, f is the Fermi–Dirac distribution function and G_i is the single-particle Green's function, related to the broadened DOS of the ith Landau level via

$$D_i = \frac{1}{2\pi\lambda^2}\left(-\frac{1}{\pi}\mathrm{Im}\,G_i\right).$$

Therefore D_i takes different functional forms depending on whether Lorentzian or Gaussian broadening of the Landau levels is assumed. Equation (6) represents the conductivity for a single layer; however, as $W \ll \hbar\omega_c$, the three-dimensional conductivity can be obtained by scaling with the lattice parameter b. To a first approximation [19], the off-diagonal Hall component can be obtained via the integration

$$\sigma_{xy,2D} = -\sigma_{yx,2D} = -\frac{e}{B}\int f\sum_i D_i\,\mathrm{d}E. \tag{7}$$

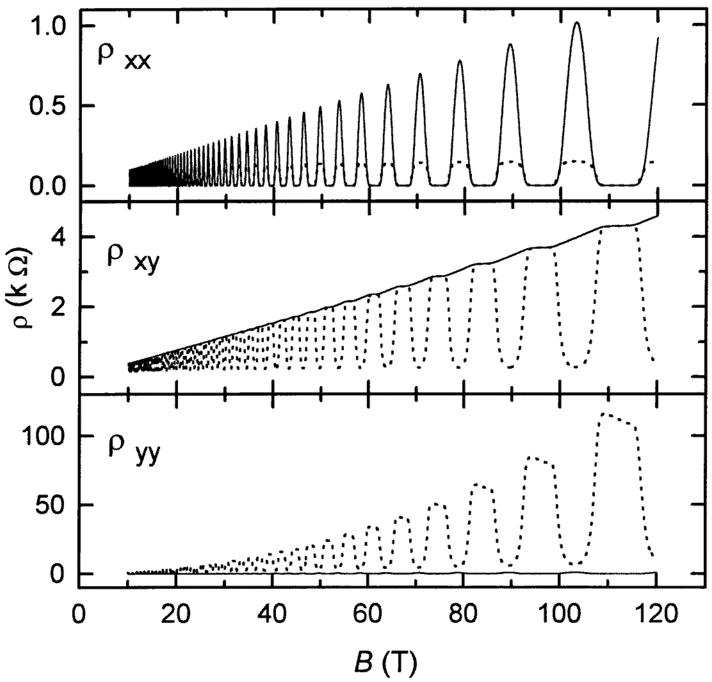

Figure 2. Calculated values of ρ_{xx} (top section) ρ_{xy} (centre section) and ρ_{yy} (bottom section) assuming Drude-like quasi-one-dimensional conductivity (dashed line) and localized quasi-one-dimensional states (solid line). For other parameters, see the text. Note that quantum Hall plateaux are observed in ρ_{xy} in both cases.

The chemical potential μ is determined by keeping the integral

$$N_{1\mathrm{D}} + N_{2\mathrm{D}} = \int \left(D_{1\mathrm{D}} + \sum_i D_i \right) f \ \mathrm{d}E \tag{8}$$

over the total number of states constant [6], where $D_{1\mathrm{D}}$ is the density of quasi-one-dimensional states. As a consequence, the carriers flow continually back and forth between the quasi-two-dimensional and quasi-one-dimensional Fermi-surface sections, the latter acting as a charge reservoir (see figure 1 of reference [6]). Note that the field-averaged DOS of the quasi-one-dimensional Fermi-surface section $D_{1\mathrm{D}}$ has recently been estimated to be 0.4 times that of the quasi-two-dimensional Fermi-surface section, $D_{2\mathrm{D}}$, in the α-phase BEDT-TTF salts [6, 20].

Figure 2 (dashed lines) shows ρ_{xx}, ρ_{yy} and ρ_{xy} calculated using the above method with the parameters $m^* = 2.5m_e$, $F = 670$ T and $\tau^{-1} = 0.2 \times 10^{12}$ s^{-1}, appropriate for α-(BEDT-TTF)$_2$MHg(SCN)$_4$ (M = K, Tl) at high magnetic fields [6]; Lorentzian broadening of the Landau levels has been assumed [21]. There are two important contrasts with the familiar magnetoresistance data from the two-dimensional semiconductor systems [2]. Firstly, the oscillations in ρ_{xx} and ρ_{yy} differ in phase, amplitude and general appearance (compare the upper and lower sections of figure 2); in the semiconductor systems, ρ_{xx} and ρ_{yy} usually behave identically [22]. Secondly the Hall resistance, ρ_{xy} (see figure 2, centre section), exhibits oscillatory behaviour between the plateaux caused by the influence of

$\sigma_{xx,1D}$ within the conductivity tensor. In spite of these oscillations, quantum Hall plateaux occur when μ is situated directly between Landau levels.

The flatness of the quantum Hall plateaux in figure 2 depends on the magnitude of the residual conductivity $\sigma_{xx,2D} = \delta\sigma$ of the quasi-two-dimensional Fermi-surface section when μ is situated in the gap in the DOS. It can be shown that in the limit $E' > \hbar\tau^{-1}$, where E' is the energy relative to the centre of the Landau level (i.e. the energy away from the centre of the Landau level), the real part of the Green's function becomes $\mathrm{Re}\, G_i \approx 1/E' > \mathrm{Im}\, G_i$ [19]. This result is essentially independent of the type of the Landau-level broadening. Hence it can be shown that

$$\delta\sigma \sim \frac{ie^2}{\hbar} \frac{\omega_c \tau}{8} e^{-\pi\omega_c\tau/8} \tag{9}$$

for Gaussian broadening of the Landau levels or

$$\delta\sigma \sim \frac{ie^2}{\hbar} \frac{1}{(\pi\omega_c\tau)^2} \tag{10}$$

for Lorentzian broadening. In both cases, the residual conductivity is very much less than the Hall conductivity. Hence, assuming that $\delta\sigma$ is small, the magnetoresistivity in the vicinity of the Hall plateau is

$$\begin{vmatrix} \rho_{xx} & \rho_{xy} \\ \rho_{yx} & \rho_{yy} \end{vmatrix} \approx \begin{vmatrix} R_H^2 \delta\sigma & R_H \\ -R_H & R_H^2(\sigma_{1D,0} + \delta\sigma) \end{vmatrix} \tag{11}$$

where R_H is now $B/(e(N_{2D} + \delta N))$ and where $\delta N = \gamma_{an} N_{1D}$. The most important effect of the anisotropy of the quasi-one-dimensional Fermi-surface component is therefore that of increasing the value of the Hall resistance at the plateau by an amount proportional to γ_{an} (see equation (4)). Since the quasi-one-dimensional Fermi surface is extremely anisotropic (see the above estimates of γ_{an}), this additional component is vanishingly small in figure 2.

In the preceding text we have shown that the plateaux in ρ_{xy} which characterize the quantum Hall effect are a natural phenomenon in a system containing both quasi-two-dimensional and quasi-one-dimensional carriers, without the need for localized states. In this sense, the dashed line of figure 2 represents the most unfavourable case; i.e. the conductivity of the quasi-one-dimensional states remains Drude-like even at high magnetic fields. If some form of localization affects the quasi-one-dimensional carriers at high magnetic fields, then there is an increased probability of observing resistivity behaviour more like that seen in the two-dimensional semiconductor systems. It is known that the conductivity of quasi-one-dimensional Fermi-surface sheets becomes increasingly one-dimensional as B increases [14]. At a certain threshold field B_c determined by the condition $\hbar\omega_c > 2t_c$, the quasi-one-dimensional carriers become effectively confined to a single chain of BEDT-TTF molecules [14, 16]. According to the estimates based on the band-structure calculations described above, we should expect this one-dimensionalization to occur at ∼10–100 T. Under such 1D confinement, the quasi-one-dimensional carriers become highly susceptible to localization effects at low temperatures [16]. A detailed calculation of the electron transfer between BEDT-TTF molecules at high magnetic fields is beyond the scope of the present letter. Nevertheless for the purpose of making a comparison, we have additionally calculated the magnetoresistance for the case where the quasi-one-dimensional states are completely localized (figure 2, solid lines). When this localization occurs, the quantum Hall effect, ρ_{xx} and ρ_{yy} resemble those observed in the two-dimensional semiconductor systems [2]. Realistically, we might expect the real magnetoresistance to lie somewhere between these two extreme limits.

Returning to the calculations made for the case where the quasi-one-dimensional states are assumed to remain Drude-like (figure 2, dashed lines), it is clear that ρ_{yy} exhibits prominent peaks when the minima in ρ_{xx} occur. These flat-bottomed minima in ρ_{xx} are the so-called 'ideal conducting phases' (i.e. ρ_{xx} tends to zero, resulting, under certain circumstances, in the flow of quasi-persistent currents) [21]. Clearly if this were to be the case then a quasi-persistent current of the kind observed in two-dimensional semiconductor systems in such resistivity minima (see reference [21] and references therein) could only flow in the x-direction. However, the recent experimental observation of quasi-persistent *circulating* currents at $B \sim 30$ T in α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ [4] and α-(BEDT-TTF)$_2$KHg(SCN)$_4$ [5] suggests that ρ_{yy} must also be very small in the ρ_{xx}-minima. Localization effects would greatly reduce the size of the maxima in ρ_{yy}, and in the case of strong localization (figure 2, lowest section) would cause the maxima to invert and become minima. The observation of quasi-persistent, circulating eddy currents [4, 5] therefore strongly suggests that localization of the quasi-one-dimensional states occurs at magnetic fields $\sim$30 T in at least some of the α-phase BEDT-TTF salts.

In this letter we have shown that the combined existence of a weakly warped quasi-one-dimensional Fermi surface and a quasi-two-dimensional Fermi surface can give rise to the quantum Hall effect in high magnetic fields; the existence of localized states is not necessary. However, a comparison of the calculations in this letter with recent experimental data suggests that field-induced localization of quasi-one-dimensional carriers may in fact occur in some of the α-phase BEDT-TTF salts.

This work was supported by the Belgian National Science Foundation, the EPSRC (UK) and the Royal Society (UK). We should like to thank Markus Honold, Andrew House, Dr William Hayes, Dr Stephen Blundell and Professor David Shoenberg for very helpful discussions.

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Dupuis N and Montambaux G 1992 *Phys. Rev.* B **46** 9603

[17] The fermiological expression using the experimentally determined effective mass of $2.5m_e$ yields $v_F \sim 0.7 \times 10^5$ m s^{-1} and hence $t_c \sim 2.1$ meV. This is therefore more representative of the real Fermi velocities in these materials. The large disparity between these two estimates is likely to be due to effective-mass-renormalization effects which are not taken into account in the band-structure calculations. See e.g.

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for a detailed discussion of this point.

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[22] Note that the oscillations in ρ_{xx} in figure 2 would be in antiphase with oscillations in ρ_{zz}. This point is covered in some detail in reference [6].