Quantum magneto-optics of graphite with trigonal warping

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The optical conductivity of graphite in quantizing magnetic fields is studied. Both the dynamical conductivities, longitudinal as well as Hall’s, are analytically evaluated. The conductivity peaks are explained in terms of electron transitions. We have shown that the trigonal warping in graphite can be considered within the perturbation theory at the strong magnetic field larger than 1 T approximately. The main optical transitions obey the selection rule with $\Delta n = 1$ for the Landau number $n$, however the $\Delta n = 2$ transitions due to the trigonal warping with the small probability are also essential. The Kerr rotation and reflectivity in graphite in the quantizing magnetic fields are calculated. Parameters of the Slonczewski–Weiss–McClure model are used in the fit taking into account the previous $d$HvA measurements and correcting some of them for the case of the strong magnetic fields.

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I. INTRODUCTION

Properties of graphite have attracted much attention for more than 50 years. Many of that properties were successfully explained within the Slonczewski–Weiss–McClure (SWMC) theory. The most accurate method to study the band structure of graphite is a study of the Landau levels (LLs) through experiments such as magneto-optics and magneto-transport. However, the interpretation of the experimental results involves a significant degree of uncertainty since, as it is not clear how the resonances should be identified and which electron transitions they correspond to.

The SWMC theory requires the use of many tight-binding parameters and provides the simple description of observed phenomena either in the semiclassical limit of weak magnetic fields or for high frequencies when the largest tight-binding inter-layer parameter $\gamma_1$ plays the leading role. It is more difficult to take into account the parameter $\gamma_2$ known as “trigonal warping”. Usually, it is either neglected or considered numerically. The Bohr-Sommerfeld quantization condition was applied in Ref. to find in low magnetic fields the level structure including the trigonal warping. In any case, only the problem of levels was considered so far, and no calculations of conductivities were done in order to evaluate the optic properties of graphite. The problem appearing for three-dimensional systems in the magnetic field connects partly with integrating over the momentum projection $k_z$ along the magnetic field.

The SWMC model can be simplified assuming that only the integration limits such as the $K$ and $H$ points in the Brillouin zone produce the main contributions. Such an approximation is similar to the theory of magneto-optical effects in topological insulators and graphite. However, the band extrema or the integration limits at the Fermi level can also contribute into the absorption. Therefore, the analytical expression for the dynamic conductivity in the presence of magnetic fields is needed for the interpretation of the magneto-optics experiments.

In this paper, motivated by the experimental study of the Faraday rotation in single- and multilayer graphene, we propose a theory of magneto-optics phenomena in graphite in strong magnetic fields including the interlayer hopping parameters $\gamma_3$ and $\gamma_4$ in the Hamiltonian. The trigonal warping $\gamma_1$ is considered as a perturbation with the help of the Green’s function method. Not only the energy-level structure corrected due to the trigonal warping is found, but the expressions for the longitudinal and Hall dynamical conductivities are derived. Our main theoretical finding is the reflectivity and the Kerr angle for graphite in strong magnetic fields.

II. LANDAU LEVELS IN GRAPHITE WITH TRIGONAL WARPING

Taking into account the tight-binding parameters of the SWMC theory, the effective Hamiltonian near the $KH$ line of the Brillouin zone in graphite writes in the form of Ref. where $k_{\pm} = \mp ik_x - k_y$ are the momentum components and $v$ is the velocity parameter in the intra-layer directions; $\tilde{\gamma}_j$ are the functions of the $k_z$ momentum in the main axis direction

\[
\tilde{\gamma}_2 = 2\gamma_2 \cos (2k_z d_0), \quad \tilde{\gamma}_5 = 2\gamma_5 \cos (2k_z d_0) + \Delta, \quad \tilde{\gamma}_i = 2\gamma_i \cos (k_z d_0) \quad \text{for} \quad i = 1, 3, 4,
\]

with the distance $d_0 = 3.35$ Å between the layers in graphite. The nearest-neighbor hopping integral $\gamma_0 \approx 3$ eV corresponds with the in-layer inter-atomic distance $\gamma_0 \approx 3$ eV.
where the band number $n$ is found, they are shown in Fig. 1 as a function of the magnetic fields.

We obtain a system of the linear equations for the eigenvalues involving the creation $a^+$ and annihilation operators $a$.

We write only one of two space coordinates including the corresponding degeneracy proportional to the magnetic field in the final results.

We search the eigenfunction of the Hamiltonian (1) in the form

$$\psi_{sn}^{\alpha}(x) = \begin{cases} C_{sn}^{1}\varphi_{n-1}(x) \\ C_{sn}^{2}\varphi_{n}(x) \\ C_{sn}^{3}\varphi_{n+1}(x) \\ C_{sn}^{4}\varphi_{n+2}(x) \end{cases},$$

where $\varphi_n(x)$ are orthonormal Hermitian functions with the Landau numbers $n \geq 0$. One sees that every row in the Hamiltonian (1) becomes proportional to the definite Hermitian function if the terms with $\gamma_3$ is omitting. We will show that the terms proportional to $\gamma_3/\gamma_0$ can be considered within the perturbation theory at strong magnetic fields.

Canceling the Hermitian functions from the equations, we obtain a system of the linear equations for the eigenvector $C_{sn}$

$$
\begin{pmatrix}
\gamma_5 - \varepsilon & \omega_c \sqrt{n} & \gamma_1 \\
\omega_c \sqrt{n} & \gamma_2 - \varepsilon & \omega_4 \sqrt{n-1} \\
\gamma_1 & \omega_4 \sqrt{n} & \gamma_6 - \varepsilon \\
\omega_4 \sqrt{n-1} & 0 & \gamma_2 - \varepsilon
\end{pmatrix}
\begin{pmatrix}
C_{sn}^{1} \\
C_{sn}^{2} \\
C_{sn}^{3} \\
C_{sn}^{4}
\end{pmatrix} = 0
$$

where the band number $s = 1, 2, 3, 4$ numerates the solutions at given $n$ from the bottom, $\omega_c = v\sqrt{2|\varepsilon|B/c}$ and $\omega_4 = \gamma_4/\gamma_0$.

The eigenvalues of the matrix in Eq. (3) are easily found, they are shown in Fig. 1 as a function of the momentum $k_z$. For each Landau number $n \geq 2$ and momentum $k_z$, there are four eigenvalues $\varepsilon_s(n)$ and four corresponding eigenvectors, Eq. (2), marked by the band subscript $s$. We will use the notation $|sn\rangle$ for levels. In addition, there are four levels. One of them,

$$\varepsilon_1(n = 0) = \tilde{\gamma}_2$$

for $n = 0$ with the eigenvector $C_0 = (0, 1, 0, 0)$ as is evident from Eq. (2). It intersects the Fermi level and belongs to the electron (hole) band near the $K$ (H) point. Others three levels indicated with $n = 1$ and $s = 1, 2, 3$ are determined by first three equations of the system (3) with $C_{s1}^1 = 0$. The $|21\rangle$ level is close to the $|10\rangle$ level. In the region of $k_z$, $\gamma_1/\cos z \gg \gamma_2$, where the electrons are located, this level has the energy

$$\varepsilon_2(n = 1) = \tilde{\gamma}_2 - 2\omega_4^2/\gamma_1^2 \gamma_0^2.$$ 

In the same region, the two closest bands ($s = 2, 3$) with $n \geq 2$ are written as

$$\varepsilon_{2,3}(n) = \tilde{\gamma}_2 - \omega_4^2/\gamma_1^2 (2n - 1) + \omega_4^2/\gamma_1 \sqrt{n(n - 1)}$$

within accuracy of $(\gamma_4/\gamma_0)^2$.

A simplest way to evaluate the corrections resulting from the warping $\gamma_3$ consists in the consideration of the relative Green’s function having the poles at the electron

| Parameter | Values |
|-----------|--------|
| $a_0$     | 1.415 Å |
| $v$       | 1.5a_0 \gamma_0 = 10^6 m/s |

For the zero magnetic field, the eigenvalues of the Hamiltonian give four close bands. In the magnetic field $B$, the momentum projections $k_{x,y}$ become the operators obeying the commutation rule $[\hat{k}_+, \hat{k}_-] = -2\varepsilon hB/c$, and we use the relations

$$\hat{k}_+ = \sqrt{2|\varepsilon|hB/c}, \quad \hat{k}_- = \sqrt{2|\varepsilon|hB/c} a^+,$$

TABLE I: The parameters of the Hamiltonian, Eq. (1), their values in the SWMC model, and obtained in the experimental works, all in meV.

|      | $\gamma_0$ | $\gamma_1$ | $\gamma_2$ | $\gamma_3$ | $\gamma_4$ | $\Delta$ | $\varepsilon_F$ |
|------|------------|------------|------------|------------|------------|----------|--------------|
| SWMC | 3050       | 360        | -10.2      | -270       | -150       | -1.5     | -4.1         |
| $\gamma_0$ | 3160       | 390        | 276        | 44         | 38         | 8        | -24          |
| $\gamma_0$ | 3120       | 380        | -21        | 315        | 120        | -3       | -2           |

$^a$SWMC, Ref. 6, $^b$Mendez et al, Ref. 6, $^c$Doezema et al, Ref. 6.

| $\omega_1$ | $\omega_2$ | $\omega_3$ | $\omega_4$ |
|------------|------------|------------|------------|
| 0.15       | 0.2        | 0.15       | 0.15       |

FIG. 1: (Color online) LLs $\varepsilon_{sn}$ for $n = 0$ to 4 in four bands $s = 1, 2, 3, 4$ (in dotted, solid, dashed, and dash-dotted lines, respectively) as functions of momentum $k_z$ along the $KH$ line in the Brillouin zone ($K = 0$, $H = \pi/2d_0$) at the magnetic field $B = 7$ T with the SWMC model parameters given in Tabl. 1. The main electron transitions shown in the right panel below 100 meV are possible between the levels with the selection rule $\Delta n = \pm 1$, see text.
levels. The corrections to the levels can be found in the iterations

$$G_{m+1}(x,x') = \int dx'' G_0(x,x'')V(x'')G_m(x'',x'),$$

where $V(x)$ has only two matrix elements $V^{42} = \omega_0^2 \alpha_{+}/\gamma_0$ and $V^{24} = V^{42*}$ in the Hamiltonian $H$. The Green’s function of the unperturbed Hamiltonian writes

$$G^0_{\alpha\beta}(\varepsilon,x,x') = \sum_{sn} \frac{\psi^\alpha_{sn}(x)\psi^\beta_{sn}(x')}{\varepsilon - \varepsilon_{sn}}.$$

In the second iteration, we get the corrections

$$\int dx_1 dx_2 G^0_{\alpha\beta}(x_1,x_2) V^{42}(x_1) G^{22}(x_1,x_2) V^{24}(x_2) G^0_{\alpha\beta}(x_2,x')$$

and the similar term with the substitution of the superscripts $2 \leftrightarrow 4$. The matrix elements of the perturbation $V$ are easily calculated with respect to the Hermitian functions of Eqs. (7, 8) and we obtain for the diagram shown in the upper part of Fig. 2

$$\left(\frac{\omega_0^2 \gamma_0}{\gamma_0}\right)^2 \sum_{s',sn} \frac{(n-2)|C^4_{sn} C^{s',n-3}_{s',n-3}|^2 \psi^\alpha_{sn}(x)\psi^\beta_{sn}(x')}{(\varepsilon - \varepsilon_{sn})(\varepsilon - \varepsilon_{s',n-3})(\varepsilon - \varepsilon_{s,n})}.$$

This correction plays an important role near the poles of the Green’s function. Therefore, we can substitute $\varepsilon_{sn}$ instead of $\varepsilon$ in the second factor of the denominator and represent this correction as a shift $\delta \varepsilon_{sn}$ of the poles $(\varepsilon - \varepsilon_{sn} - \delta \varepsilon_{sn})^{-1}$ with

$$\delta \varepsilon_{sn}(n) = \left(\frac{\omega_0^2 \gamma_0}{\gamma_0}\right)^2 \sum_{s',n} \left\{ \frac{(n-2)|C^4_{sn} C^{s',n-3}_{s',n-3}|^2}{\varepsilon_{s'}(n) - \varepsilon_{s'}(n-3)} ight. + \left. \frac{(n+1)|C^2_{sn} C^{s',n+3}_{s',n+3}|^2}{\varepsilon_{s'}(n) - \varepsilon_{s'}(n+3)} \right\},$$

where the first term should be omitted for $n < 0$. In fact, our illustration is nothing but a calculation of the electron self-energy and the naive expansion of the denominator can be indeed replaced by summing up the corresponding diagrams.

Comparing the corrections, Eq. (9), with the main contribution Eq. (5), we find that the perturbation theory is valid when an expansion parameter $(\gamma_3 \gamma_1/\gamma_0 \omega_0)$ becomes small, i.e. for the strong magnetic fields $B > 1T$. The corrected level writes

$$\varepsilon_1(n = 0) = \varepsilon_2 + \left(\frac{\omega_0 \gamma_0}{\gamma_0}\right)^2 \sum_{s'} \frac{|C^4_{sn}|^2}{\gamma_2 - \varepsilon_{s'}(3)}.$$

The (21)–level is very close to the level with $n = 0$, Eqs. (4) and (10).

Our expressions for the levels with the corrections (9) and (10) give the same results as obtained in Ref. 21, by the numerical method of truncating the infinite matrix.

III. CONDUCTIVITIES IN MAGNETIC FIELDS

In the collisionless limit when the relaxation rate $\Gamma$ is much less than the frequency, $\Gamma \ll \omega$, the conductivity is expressed in terms of the correlation function

$$\mathcal{P}(\omega) = T \sum_{\omega_m} \int dx dx' Tr \left\{ \psi^\dagger G(\omega_+ + x + x') \psi^\dagger G(\omega_- + x', x) \right\}$$

where we should (i) summarize over Matsubara’s frequencies $\omega_m$, (ii) take the trace over the Landau and band numbers, (iii) make an analytic continuation into real frequencies $\omega$, and (iii) substrate from the result its value at $\omega = 0$ (for details see Ref. 22).

The velocity matrices $v^\dagger$ in Eq. (11) are given by the derivative of the Hamiltonian, Eq. (10).

$$v = \frac{\partial H(k)}{\partial k}.$$

First we consider the largest velocity operators, Eq. (12), which do not involve the parameter $\gamma_3/\gamma_0$. Straightforward calculations yield two independent components of the dynamical conductivity

$$\{ \sigma_{xx}(\omega), i \sigma_{xy}(\omega) \} = i \sigma_0 \frac{4\omega^2}{\pi^2} \sum_{n,s,s'} \int dz \Delta_{ss'n} |dz'n| \left[ \Delta_{ss'n} \right]$$

where $\Delta_{ss'n} = \varepsilon_{sn} - \varepsilon_{s'n} + 1$ is the level spacing including the corrections given in Eqs. (9) and (10), $\Delta f_{ss'n} = f(\varepsilon_{sn+1}) - f(\varepsilon_{sn})$ is the difference of the corresponding Fermi-Dirac functions and

$$d_{ss'n} = C^1_{sn} C^1_{s'n} + C^3_{sn} C^1_{s'n} + C^1_{sn} C^3_{s'n} + C^3_{sn} C^3_{s'n}$$

is the dipole matrix element. These electron transitions obey the selection rule

$$\Delta n = 1,$$
layers within the distance unit in the main axis-direction. Eq. (11) we get an additional term in the conductivity, instead of the matrix element given by Eq. (14). This transitions obey the selection rule

$$\Delta n = 2$$

and will be referenced as the strong lines. The integration over the Brillouin half-zone, $0 < z < \pi/2$, and the summation over the Landau number $n$ as well as the bands $s, s'$ should be done in Eq. (13). The conductivity units

$$\sigma_0 = \frac{e^2}{4\hbar d_4}$$

have a simple meaning, being the graphene dynamic conductivity multiplied by the number $1/d_4$ of layers within the distance unit in the main axis-direction.

Now we consider the terms with $\gamma_3/\gamma_0$ in the velocity operators, Eq. (12). Calculating the correlation function Eq. (11) we get an additional term in the conductivity, which can be obtained from Eq. (11) with the substitutions

$$n + 1 \rightarrow n + 2$$

and with the matrix element

$$d_{ss'n} = (\gamma_3/\gamma_0)C_{sn}C_{s'n+2}$$

instead of the matrix element given by Eq. (14). This transitions obey the selection rule

$$\Delta n = 2$$

and will be referenced as the weak lines.

So far we considered the $\gamma_3$ corrections to the Green’s function, i.e. to the levels. However, there are so-called vertex corrections to the self-energy shown at the bottom of Fig. 2. They are resulted from the quartet of the coupled Landau levels, which interfere while the selection rules $\Delta n = 1$ and $\Delta n = 2$ are allowed. For compactness, let us denote this quartet of given $\Delta n = 2$ are allowed. For compactness, let us denote this quartet of given as following

$$a = |sn|, b = |s'|, c = |s_1, n + 3|, d = |s'_1, n + 4|$$

where the band numbers $s, s', s_1$, and $s'_1$ are arbitrary.

The corresponding corrections to conductivities write

$$\frac{d\sigma_{xx} \omega)} i d\sigma_{xy}(\omega) = 2i\sigma_0 \sum_{nss's_1s_1'} \int_0^{\pi/2} \frac{2\omega^2 \gamma_0}{\gamma_0 \Delta n} \left( C_{ss_1} C_{s'n+2} \right)$$

and

$$d_{ss'n} = (\gamma_3/\gamma_0)C_{sn}C_{s'n+2}$$

instead of the matrix element given by Eq. (14). This transitions obey the selection rule

$$\Delta n = 2$$

and will be referenced as the strong lines. The integration over the Brillouin half-zone, $0 < z < \pi/2$, and the summation over the Landau number $n$ as well as the bands $s, s'$ should be done in Eq. (13). The conductivity units

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have a simple meaning, being the graphene dynamic conductivity multiplied by the number $1/d_4$ of layers within the distance unit in the main axis-direction.
The positions of the lines for fields in the range of 10 – 30 T agree with observations of Refs. 8,15.

The optical Hall conductivity $\sigma_{xy}(\omega)$ in the ac regime is shown in Fig. 3 (a) and (b). The conductivities $\sigma_{xx}(\omega)$ and $\sigma_{xy}(\omega)$ allow us to calculate the Kerr rotation and the reflectivity as functions of frequency [see Fig. 3 (c) and (d)]. It is evident that the interpretation of the Kerr rotation governed by the conductivity $\sigma_{xy}(\omega)$ is much more complicated in comparison with the longitudinal conductivity.

The Kerr angle and reflectivity shown in Fig. 4 for the different magnetic fields demonstrate the strong field dependence of the magneto-optic phenomena.

V. SUMMARY AND CONCLUSIONS

In conclusions, we have evaluated the perturbation theory for the matrix Hamiltonian, which permits to calculate the corrections to the eigenvalues resulting from the small matrix elements particularly from the trigonal warping. We have shown that the trigonal warping in graphite can be considered within the perturbation theory at the strong magnetic field larger than 1 T approximately. We have found that the main optical transitions obey the selection rule $\Delta n = 1$ for the Landau number $n$, however the $\Delta n = 2$ transitions due to the trigonal warping with the small probability are also essential. The good agreement between the calculations and the measured Kerr rotation and reflectivity in graphite in the quantizing magnetic fields is achieved. The SWMC model parameters are used in the fit taking into account the previous dHvA measurements and correcting the Fermi energy for the case of the strong magnetic fields.

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