High-frequency conductivity of multilayer graphene and graphite under the conditions of quantum cyclotron resonance

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The conductivity tensor of a layered conductor with the Dirac-type energy spectrum of charge carriers placed in a quantizing magnetic field under the condition of normal skin-effect is investigated using the method of quantum kinetic equation. It is shown that under the cyclotron resonance conditions there appear high-temperature quantum oscillations of conductivity, which are weakly sensitive to thermal broadening of the Fermi level. We present the expressions for the classical and high-temperature contributions to the conductivity tensor which determine the conductivity in the range of not too low temperatures where the Shubnikov–de Haas oscillations are vanishing. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4884523]

1. Introduction

Increased interest in graphene,[1] which is fully justified by the unusual physical properties of this material, has resulted in the emergence of new experimental and theoretical studies of physically similar materials, in particular, such as multi-layered graphene and well-known graphite.[2,3] This makes the studies of high-frequency characteristics of these materials highly relevant. Thus, in Ref. 4, the Lifshitz transition in graphite has been experimentally observed using cyclotron resonance and theoretically described. In Ref. 5 the results of measurements of the cyclotron resonance in pyrolytic graphite in the geometry similar to the discussed in this paper have been published.

Linear energy spectrum of charge carriers is characterized by a substantially uneven spacing under magnetic quantization. If the cyclotron frequency depends strongly on the number of the Landau cylinder and weakly on the momentum projection on the magnetic field direction, high-temperature quantum oscillations (HTQO) of the kinetic coefficients become possible, which are rather insensitive to thermal broadening of the Fermi step. The appearance of the HTQO of the kinetic coefficients under the cyclotron resonance conditions was first predicted in Ref. 6 in normal metals. However, rather stringent conditions required for the emergence of HTQO make them unlikely to appear in conventional conductors. At the same time the HTQO might naturally appear in graphite and related materials for the typical values of parameters characterizing the electron energy spectrum.

2. Formulation of the problem

Let us consider a layered conductor with the energy spectrum of charge carriers in the form

$$\epsilon(p) = v_0(\sigma_x p_x + \sigma_y p_y) - t\cos\left(\frac{ap_z}{\hbar}\right), \qquad (1)$$

where $\hbar$ is the Planck constant, a is the distance between the layers, v_0 is the speed of conduction electrons within the layer, t is the overlap integral for the wave functions of electrons belonging to adjacent layers, σ_i is the Pauli matrix. This model has been proposed in Ref. 7 and takes into account such essential features of graphite as the linear dependence of energy on momentum in the plane of the layers in a certain spectral region and strong anisotropy of its electronic properties. An important advantage of the model (1) is its simple form, convenient for analytical description of the conductor properties. Equation (1) differs significantly from the dependence of the energy of conduction electrons in graphite proposed in the model by Slonczewski, Weiss, and McClure.[8,9] In particular, the model does not account for small in-plane anisotropy within the layers, which results in additional harmonics appearing in the cyclotron resonance.[4,5] The effects associated with the anisotropy within the layers are not the considered in this paper.

Let us assume that the quantizing magnetic field $\mathbf{B}_0 = (0,0,B_0)$ is oriented along the normal to the surface of the conductor, which is parallel to the layers. Let us consider the current response of the conductor to the field of a circularly polarized electromagnetic wave $E^{\pm} = E_x \pm iE_y$ with the frequency ω under the conditions of normal skin effect. We assume that the following inequality holds

$$\omega\tau \gg 1, \quad t \pm \varepsilon_F \gg \hbar\omega. \qquad (2)$$

We limit ourselves to the case of not too low temperatures, at which the following inequalities hold

$$T \gtrsim \hbar\Omega_{e,h}, \quad \Omega_e = \frac{v_0^2 eB}{c(t+\varepsilon_F)}, \quad \Omega_h = \frac{v_0^2 eB}{c(t-\varepsilon_F)}. \qquad (3)$$

Here, $\Omega_{e,h}$ is the cyclotron frequency at the extreme cross sections of the electron and hole regions of the Fermi surface, respectively. In this temperature range, the usual

Shubnikov-de Haas (SdH) oscillations are strongly suppressed, and the electric conductivity, Eq. (1), is determined by the classical contribution and the HTQO of conductance arising under the conditions of quantum cyclotron resonance.[6] The oscillations of this type are possible in samples with high $\Omega\tau$, which may be justified due to rather high values of the cyclotron frequency in graphite.

To calculate the current response, the quantum kinetic equation in the τ-approximation is commonly applied. There are a number of quantum oscillation effects which are very sensitive to the dissipation mechanism. Thus, the scattering by impurities can determine the amplitude of the quantum oscillations of kinetic coefficients[10] and also makes the HTQO possible in the SdH effect in layered conductors, as has been described in Refs. 11–13. These oscillations disappear in the collisionless limit. However, the HTQO considered in this paper, which arise under the conditions of quantum cyclotron resonance, exhibit the maximum magnitude in the collisionless limit, and the nature of the scattering does not affect the mechanism of the oscillation.

3. Conductivity tensor

Let us write the quantum kinetic equation in the form

$$\left[-i\omega+\frac{i}{\hbar}(\varepsilon_\nu-\varepsilon_\mu)+\frac{1}{\tau}\right]\rho^1_{\nu\mu}=-\frac{\rho^0_\nu-\rho^0_\mu}{\varepsilon_\nu-\varepsilon_\mu}eEv_{\nu\mu}, \quad (4)$$

where e is the electron charge, E is the electric field, $v_{\nu\mu}$ are the matrix elements of the charge-carriers velocity in the proper representation of the energy operator (1), and $\rho^0_\nu=\rho^0(\varepsilon_\nu)$ is the Fermi-Dirac function. After substituting the values of $\rho^1_{\nu\mu}$ in the equation for the current

$$j^i=e\ \mathrm{Sp}\left[\hat{v}^i;\hat{\rho}^1\right] \quad (5)$$

we apply the well-known identity

$$F(\hat{Z})=\int F(x)\delta(x-\hat{Z})dx,$$

similar to Ref. 10 (see Eqs. (8) and (9) in Ref. 10). After elementary transformations, the expression for the conductivity tensor can be written in the form, which is not coupled to the proper representation of the energy operator (1)

$$\sigma_{ij}(\omega)=\frac{1}{i\pi}\int\limits_{-\infty}^{\infty}\frac{d\omega'}{\omega'-\omega-i/\tau}\Phi_{ij}(\omega'), \quad (6)$$

where

$$\Phi_{ij}(\omega)=2\pi e^2\hbar\int dE\left(-\frac{\rho^0(E+\hbar\omega)-\rho^0(E)}{\hbar\omega}\right)\times\ \mathrm{Sp}\left[\hat{v}^j\delta(E-\hat{\epsilon}+\hbar\omega)\hat{v}^i\delta(E-\hat{\epsilon})\right]. \quad (7)$$

Here

$$\delta(E-\hat{\epsilon})=\frac{i}{2\pi}\left[\hat{G}^+(E)-\hat{G}^-(E)\right]$$

and

$$\hat{G}^\pm(E)=(E-\hat{\epsilon}\pm i\delta)^{-1}$$

is the one-electron Green's function. The function $\Phi_{ij}(\omega)$ determines the correlation function in the absence of scattering.[14] The one-electron Green's function for the energy spectrum of the charge carriers in the form of Eq. (1) can be conveniently written as

$$\hat{G}^\pm(E)=\frac{E+t\cos(ap_z/\hbar)+v_0\hat{R}}{[E+t\cos(ap_z/\hbar)\pm i\delta]^2-v_0^2\hat{R}^2},$$
$$\hat{R}=\hat{\sigma}_x\hat{p}_x+\hat{\sigma}_y\hat{p}_y. \quad (8)$$

It can be shown that the contribution $v_0\hat{R}$, which contains the Pauli matrices and is off-diagonal in quasi spin indices, can be neglected when the conductivity tensor is calculated for the groups of charge carriers characterized by the high-n Landau cylinders. As follows from the commutation relations for the Pauli matrices, $\hat{R}^2=\hat{p}_x^2+\hat{p}_y^2+(eB\hbar/c)\hat{\sigma}_z$. It is well-know that the eigenvalue problem for the operator of kinematic momentum squared $\hat{S}=\pi(\hat{p}_x^2+\hat{p}_y^2)$ in a magnetic field leads to the quantization rule: $s_n=2\pi(eB\hbar/c)(n+1/2)$, where n is the Landau cylinder number, $\mathbf{p}=\mathbf{P}-\frac{e}{c}\mathbf{A}$, and the vector potential $\mathbf{A}$ describes the quantizing magnetic field $\mathbf{B}_0$. The Pauli matrix $\hat{\sigma}_z$ in the denominator of Eq. (8) naturally leads to an additional shift of the quantum oscillations of kinetic coefficients due to the linear nature of the energy spectrum (the Berry phase).

After the trace over the pseudospin indices in Eqs. (6) and (7) is calculated, the expression for the circular components of the conductivity tensor $\sigma^+=\sigma_{xx}+i\sigma_{yx}$ can be written as

$$\sigma^+(\omega)=\frac{e^2v_0^2}{4\pi}\frac{8eB}{c(2\pi\hbar)^2}\sum_n\int dp_z\int dE\left[-\frac{\rho^0(E+\hbar\omega)-\rho^0(E)}{\hbar\omega}\right]\times\left[G^+_\uparrow(E+\hbar\omega)-G^-_\uparrow(E+\hbar\omega)\right]\left[G^+_\downarrow(E)-G^-_\downarrow(E)\right], \quad (9)$$

where the arrow pointing upward $\uparrow$ corresponds to the projection of pseudospin "up" with the eigenvalue of the matrix σ_z equal 1, and, respectively, -1 for a spin "down". Summation over the conventional spin is accounted by the factor of 2 in the equation.

4. Electrical conductivity of a conductor in the presence of strong thermal broadening of the Fermi level

At low temperatures, the quantum oscillations of conductivity can be determined by the contributions of the charge-carrier groups near the extreme cross sections. However, under the conditions where the temperature exceeds the energy difference between the adjacent Landau levels, the thermal broadening of the Fermi level does not necessarily results in the complete disappearance of the quantum oscillations, which can be determined by a high-temperature contribution.

Let us present the expressions describing the conductivity of a conductor at high temperatures, as per Eq. (3). In the presence of strong thermal broadening of the Fermi level, the conductivity tensor is determined by the classical and

high-temperature quantum contributions. The non-oscillating dependence of these contributions on the Fermi energy (but not on the magnetic field for the high-temperature contribution) leads to a substantial simplification of Eq. (9). Thus, it can be shown that these contributions arise only in the term of Eq. (9) which contains the product $G_{\uparrow}^{+}(E+\hbar\omega)G_{\downarrow}^{-}(E)$. Evaluation of the integral (6) for the classical part of the electrical conductivity and HTQO is reduced to the residue $\omega' - \omega - i/\tau = 0$ only. Thus, the expression for the conductivity tensor which describes the classical part and HTQO (but not the high-frequency analogue of the SdH oscillations) can be written as

$$\sigma^{+}(\omega) = \frac{e^2 v_0^2}{\pi}\frac{2eB}{c(2\pi\hbar)^2}\sum_n \int dp_z\, G_{\uparrow}^{+}(\varepsilon_F+\hbar\omega)G_{\downarrow}^{-}(\varepsilon_F). \quad (10)$$

After the Poisson formula is applied and the integrals in Eq. (10) are evaluated, the following expression for the classical part of the conductivity tensor can be obtained:

$$\sigma^{+}(\omega) = \frac{2ie^2\sqrt{(t+\varepsilon_F)(t-\varepsilon_F)}}{a\pi^2\hbar^2\omega^*} + \frac{ie^2\varepsilon_F}{2a\pi^2\hbar^2\omega^*}\left(1+\frac{\epsilon_1^2}{2\hbar\omega^*\varepsilon_F}\right)[\arccos(\varepsilon_F/t) - \arccos(-\varepsilon_F/t)]$$
$$+ \frac{ie^2(\omega^*)^2}{4\pi^2 a}\left(\frac{\epsilon_1}{\hbar\omega^*}\right)^4 \frac{1}{\sqrt{(t+\varepsilon_F)(t-\varepsilon_F)(\omega^*-\Omega_e)(\omega^*+\Omega_h)}}\,\Phi\left[\frac{(t-\varepsilon_F)^2(\omega^*+\Omega_h)}{(t+\varepsilon_F)^2(\omega^*-\Omega_e)}\right], \quad (11)$$

where

$$\Phi(x) = \operatorname{artanh}\sqrt{x} + \operatorname{artanh}\sqrt{1/x}, \quad \omega^* = \omega + i/\tau,$$
$$\epsilon_1 = v_0\sqrt{2eB\hbar/c} \quad (12)$$

is the energy difference between the zeroth and first Landau levels. Fig. 1 shows the dependence of the classical part of σ^{+} on the ratio ω/Ω_e for several values of $\Omega_e\tau$.

The jump in the real part of the conductivity at $\omega = \Omega_e$ is due to the fact that the range of cyclotron frequencies on the electron part of the Fermi surface is limited by the inequality $\Omega \geq \Omega_e$. Since the cyclotron frequency does not depend on the momentum component p_z, the resonance region of momentum space for which the following inequality holds:

$$|\omega - \Omega| \lesssim 1/\tau, \quad (13)$$

is cylindrical, and the divergence which arises near $\omega = \Omega_e$ (Fig. 1) in the collisionless limit is associated with the resonance region touching the Fermi surface at its extreme cross section.

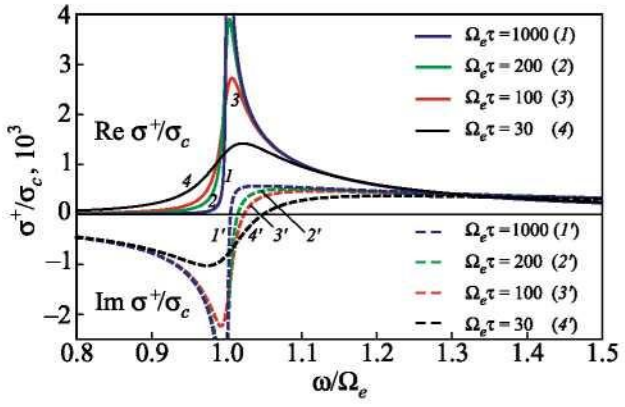

FIG. 1. Dependence of the conductivity tensor component σ^{+} (normalized by the constant $\sigma_c = e^2/(8\pi^2\alpha\hbar)$ on the ratio ω/Ω_e for several values of the relaxation time, in the case of $t/\hbar\Omega_t = 50$ and $\varepsilon_F/t = 0.25$.

The geometrical shape of the region of momentum space where the resonance condition (13) holds for charge carriers and that of the Landau cylinders coincide. Thus, the shape of the spectrum (1) is extremely favorable for the emergence of quantum conductance oscillations under the cyclotron resonance conditions. Geometric similarity of the resonance region and the contour of the quantum magnetic levels leads to the appearance of the high-temperature quantum oscillations of kinetic coefficients. Indeed, the de Haas–van Alphen (dHvA) or SdH oscillations experience strong temperature suppression of their main harmonics since their maxima are defined by the instances at which the intersection of the Fermi surface in the vicinity of the extreme sections and the Landau cylinders (taking into account the phase shift associated with the curvature of the Fermi surface near the cross sections) occurs. Therefore, thermal broadening of the Fermi level results in a broadening of the oscillation peaks. At the same time, the resonant absorption of the electromagnetic field is determined by the effective charge carriers in the region of thermal broadening of the Fermi level which simultaneously satisfy the resonance condition (13). Thus, the broadening of the effective region is limited by the resonance condition, and, for

$$T \gtrsim \frac{1}{\tau}\left(\frac{\partial\Omega}{\partial\varepsilon}\right)_{p_z}^{-1}$$

only results in widening this region in the direction p_z, while not affecting the sharpness of the oscillation peaks.

On the other hand, a significant non-equidistance of the spectrum as well as sufficiently high cyclotron frequency in graphite and related materials lead to the fact that the thickness of the cylinder "wall", which limits the resonance region, may be less than the distance between the adjacent Landau cylinders for the physically reasonable parameters of the problem.

In the case when the region of the momentum space which satisfies the resonance condition is far enough from the features of the Fermi surface, i.e. the following inequalities hold

$$|\omega - \Omega_e| \gg \hbar\Omega_e \left|\frac{\partial\Omega}{\partial\varepsilon}\right|_{p_z} \sim \frac{\hbar\Omega_e^2}{\varepsilon_F}, \quad \omega \ll v_0\sqrt{\frac{eB}{\hbar c}}, \qquad (14)$$

equation (10) leads to the following asymptotic expression that defines the high-temperature oscillations of the conductivity tensor:

$$\sigma^+(\omega) \approx \frac{e^2 t\Omega_t^2}{\pi\hbar^2 a(\omega + i/\tau)^3} \frac{1}{\sqrt{1 + \left(\dfrac{\Omega_t}{\omega + i/\tau} - \dfrac{\varepsilon_F}{t}\right)^2}} \times \sum_{k=1}^{\infty} (-1)^k \exp\left[-2\pi i k \frac{t\Omega_t}{\hbar(\omega + i/\tau)^2}\right], \qquad (15)$$

where

$$\Omega_t = \frac{eBv_0^2}{ct} = \frac{2\Omega_e\Omega_h}{\Omega_e + \Omega_h}.$$

It is easy to see that the high-temperature oscillations described by Eq. (15) are sensitive to the relaxation time τ and only possible in sufficiently pure samples. Their amplitude will not be vanishingly small if

$$\omega\tau \gtrsim \frac{eB\hbar v_0^2}{c(\hbar\omega)^2} \gg 1, \qquad (16)$$

i.e., when $\Omega\tau$ is at least comparable with the number of Landau levels below the resonance region, which is not too high in semimetals and, in particular, graphite. On the other hand, the condition (16) can be satisfied in a conductors with high cyclotron resonance frequency at the resonance condition $\omega \approx \Omega$. Graphite and multilayer graphene are characterized by a fairly high cyclotron frequency corresponding to the cyclotron masses $m \approx 0.05\ M_e$ on extreme sections, where M_e is the electron mass. In Ref. 13 the observation of cyclotron resonance with the cyclotron frequencies of the order of 0.1–0.25 eV for the groups of carriers with the Landau cylinder indices of about 10 has been reported already at magnetic fields of 0.1 T. Under such conditions, the inequality (16) will be valid already at $\hbar/\tau \lesssim 0.01$ eV.

The HTQO are weakly sensitive to thermal broadening of the Fermi step. However, at relatively high temperatures they will be suppressed due to scattering by phonons, which is particularly important for the case of the high cyclotron frequencies, as the latter implies the competition of the HTQO and low-temperature contributions at a higher temperatures. Analysis of the effect of phonon scattering is not the subject of the present work, however, following the estimates given in Ref. 6 (see the first equation in Sec. 5), the abnormally high Debye temperature of graphite should reduce the influence of phonon scattering.

Fig. 2 shows the quantum oscillations of the real part of conductivity which are associated with the resonant absorption of electromagnetic waves. Disappearance of the oscillatory dependence at high frequencies corresponds to the frequency of the electromagnetic wave matching the maximum value of the cyclotron frequency which corresponds to the transition between the zeroth and first Landau levels. Thus, the high-temperature oscillations of the conductivity tensor component σ^+ are limited to the frequency range of electromagnetic waves

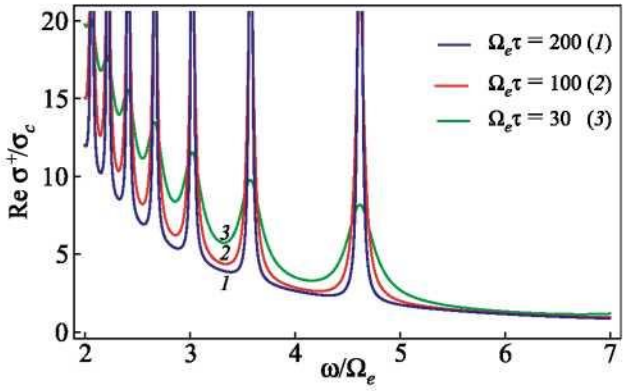

FIG. 2. High-temperature quantum oscillations of the conductivity tensor component σ^+, normalized by the constant $\sigma_c = e^2/(8\pi^2\alpha\hbar)$, for several values of the relaxation time in the case of $t/\hbar\Omega_t = 50$ and $\varepsilon_F/t = 0.25$.

$$\Omega_e \le \omega \le v_0\sqrt{\frac{2eB}{\hbar c}}. \qquad (17)$$

The oscillation peaks appear when $\hbar\omega$ matches the energy difference between the adjacent Landau cylinders, i.e., when $t\Omega_t/(\hbar\omega^2) = (n + 1/2)$, $n = 0, 1, 2, 3\ldots$

5. Conclusions

Drawing on the example of the model energy spectrum, it is shown that in layered conductors with a linear energy spectrum within the plane of the layers, quantum high-temperature conductance oscillations may appear under the conditions of cyclotron resonance. This type of conductors includes multi-layer graphene and graphite. The effect is not limited to this type of conductors, however its observation in conventional conductors is considerably more difficult, as it requires fulfilment of the criteria

$$\left|\frac{\partial\Omega}{\partial p_B}\right|_S \lesssim \frac{v_B\Delta\Omega}{T}, \quad \left|\frac{\partial\Omega}{\partial S}\right|_{p_B} \gtrsim \frac{c}{eB\hbar\tau}, \qquad (18)$$

where v_B is the projection of the velocity vector on the direction of magnetic field. The cyclotron frequency Ω is considered as a function of the projection of momentum on the direction of the quantizing magnetic field p_B, and S is the cross-sectional area in momentum space. The quantity $\Delta\Omega$ is equal to the difference of the cyclotron frequencies of the adjacent magnetic quantum levels at a fixed value of p_B. The independence of (S, p_B) on p_B in the model (1) results in the fact that the HTQO are insensitive to the thermal broadening of the Fermi level. However, in real conductors, the conditions for the emergence of HTQO will not be as favorable, and the violation of the first inequality in Eq. (18) will lead to the appearance of an exponentially strong damping of the oscillations. Inequality (16) for the model (1) and the second inequality in Eq. (18) limit the observation of HTQO to the samples of high purity.

In the materials of graphite family, the HTQO can be described in terms of the model (1) if the resonance condition $|\omega - \Omega| < 1/\tau$ is fulfilled for a group of charge carriers near the self-intersection points of the Fermi surface, where

the electron energy spectrum is close to linear. Estimates for graphite show that this condition corresponds to the cross sections of the Fermi surface, the area of which is less than $S_d \sim 10^{12}$ cm^{-2}. Good agreement of the oscillation pattern with the results of the present work is expected in the frequency range $\omega > \omega_d = \alpha B$, where $\alpha = eBv_0/\left(c\sqrt{\hbar S_d}\right) \sim 10^{13}$ Hz/T, i.e., at characteristic frequencies which are at the border of submillimeter and infrared ranges.

In the case of a linear energy spectrum, the cyclotron mass exhibits a square-root dependence on the Landau cylinder number n: $m_c \approx (\hbar/v_0)\sqrt{2n(eB\hbar/c)}$. Its value in the vicinity of the Dirac point is significantly lower than that on the extreme cross sections. Thus, the discrete nature of the energy spectrum can manifest itself near the Dirac points in the magnetic fields $\sqrt{n_F}$ times lower than those in the cases of SdH and dHvA oscillations. Here, n_F is the cylinder number corresponding to the groups of charge carriers at the extreme sections of the Fermi surface.

The stringent conditions required for the appearance of the HTQO and described by Eq. (18), make it difficult to observe the HTQO in traditional conductors. At the same time, these conditions can be met in graphite and related materials due to their quasi-2D nature and non-equidistant energy spectrum of the charge carriers. Relatively small value of the cyclotron masses in these conductors makes also possible the conditions (16) and (18), which are necessary for the emergence of these oscillations.

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