

Quantum capacitance oscillations in graphene under crossed magnetic and electric fields

Z. Z. ALISULTANOV^{1,2,3(a)} and M. S. REIS^{4(b)}

¹ *Amirkhanov Institute of Physics Russian Academy of Sciences, Dagestan Science Centre Makhachkala, Yagarskogo str., 94, 367003, Makhachkala, Russia*

² *Prokhorov General Physics Institute Russian Academy of Sciences - Vavilov Str., 38, 119991, Moscow, Russia*

³ *Dagestan State University - Gadzhiev Str., 43-a, 367000, Makhachkala, Russia*

⁴ *Instituto de Física, Universidade Federal Fluminense - Av. Gal. Milton Tavares de Souza s/n, 24210-346, Niterói-RJ, Brasil*

received 22 October 2015; accepted in final form 22 January 2016

published online 15 February 2016

PACS 81.05.ue – Graphene

PACS 84.37.+q – Measurements in electric variables (including voltage, current, resistance, capacitance, inductance, impedance, and admittance, etc.)

Abstract – Quantum oscillations of metallic systems at low temperatures are one of the key rules to experimentally access their electronic properties, such as energy spectrum, scattering mechanisms, geometry of Fermi surface and many other features. The importance of these knowledge is enormous, since from these a thorough understanding of the anomalous Hall effect, thermopower and Nernst coefficients, just to name a few, is possible; and from those knowledge, plenty of applications arise as emerging technologies. In this direction, the present contribution focuses on a complete description of quantum capacitance oscillations of monolayer and bilayer graphene under crossed electric and magnetic fields, considering to this purpose the Lifshitz-Onsager quantization condition. We found a closed theoretical expression for the quantum capacitance and highlight their amplitude, period and phase —important parameters to access the electronic properties of graphene. These results open doors for further experimental studies.

Copyright © EPLA, 2016

Magnetic and conductive thermal properties of graphene are some of the actual problems in experimental and theoretical condensed-matter physics [1]; mainly because of its two-dimensional lattice and Dirac spectrum of energy, leading thus to unique features. The magnetic field is responsible for the unusual energy spectrum of graphene [2], since its Landau levels (LLs) are nonequidistant (the gap between the first two LLs in a magnetic field of 10 T is larger than 1000 K), and then remarkable effects arise, such as the giant magnetic [3,4] and thermo-magnetic effects [5], as well as the unusual quantum Hall effect, which can be observed even at room temperature [1,6,7]; and these facts make therefore graphene a promising material for modern nanoelectronics. The oscillating magnetocaloric effect is also remarkable, since the normal and inverse effects can be tuned by the final value of applied magnetic field [8–18]; and from this

normal-to-inverse effect an enhanced thermomagnetic hysteresis cycle was proposed [19].

One key rule of the above-described properties (and even many others) is the density of states (DOS), which is linearly dependent on the energy; and for undoped graphene, the DOS vanishes at the Fermi level (corresponding thus to the Dirac point). A thorough experimental and theoretical study of this feature shall then be absolutely necessary, since from those findings, deeper and advanced technological achievements are possible. In this direction, one experimental method to study the DOS is based on the measurement of quantum oscillations of physical parameters in high magnetic field, to then access their electronic spectrum, scattering mechanisms, geometry of Fermi surface and many other features.

Based on above description, the present letter focuses on the investigation of the oscillatory properties of quantum capacitance of graphene under crossed magnetic and electric fields. Similar studies have been carried out to explore magnetization [20] and the electrocaloric effect [15]; and

^(a)E-mail: zaur0102@gmail.com

^(b)E-mail: marior@if.uff.br

all these are indeed useful for further experimental studies and then deepen our knowledge on the DOS of graphene. All of these are important challenges to overcome for a thorough understanding of the behavior of these materials and for planning new applications.

The graphene problem can be handled and solved by different theoretical tools as, for instance, an exact evaluation of Dirac's equation; but for the present letter we considered the quasi-classical approach based on the Lifshitz-Onsager quantization conditions [21,22]. We also considered the graphene sheet on the xy plane, the magnetic field as $\vec{H} = (0, 0, H)$ and the electric field as $\vec{E} = (E, 0, 0)$. From these assumptions, the quantized area enclosed by an electron trajectory in momentum space is given by [23,24]:

$$A(\epsilon') = \frac{2\pi\hbar eH}{c}(j + \gamma_{\pm}), \quad (1)$$

where j is an integer, e represents the electron charge, $c = 3 \times 10^{10}$ cm/s is the light speed and

$$\gamma_{\pm} = \gamma \pm \frac{m(\epsilon)}{2m}. \quad (2)$$

Above,

$$m(\epsilon) = \frac{1}{2\pi} \frac{dA(\epsilon)}{d\epsilon}, \quad (3)$$

is the electron cyclotron mass; γ is a constant that defines the energy of the zeroth Landau level (it assumes 1/2 for non-relativistic gas and zero for graphene); $m(\mu)/2m$ is the Zeeman splitting and m is the electron mass. Similarly to what was done before [25], in the present work we also ignored the Zeeman splitting of the Landau levels. Moreover,

$$\epsilon' = \epsilon - \vec{v}_0 \cdot \vec{p}, \quad (4)$$

where $\vec{v}_0 = cE\hat{y}/H$ is the average drift velocity of the electron perpendicularly to EH plane and

$$\epsilon = \nu_b v_F p \quad (5)$$

is the energy spectrum of a monolayer graphene (MG) in the vicinity of the Dirac point. In addition, $v_F \approx 10^8$ cm/s is the Fermi velocity and $\nu_b = \pm 1$ is the band index (“+” for the conduction band and “-” for the valence band).

From the above, it is easy to show that a curve with constant energy ϵ' is an ellipse on the $p_x p_y$ plane, with the following parameters: $a = \epsilon'/\sqrt{v_F^2 - v_0^2}$ and $b = \epsilon'v_F/(v_F^2 - v_0^2)$. Then, the quantized area on the momentum space reads as

$$A(\epsilon') = \pi ab = \frac{\pi\epsilon'^2}{v_F^2(1 - \beta^2)^{3/2}}, \quad (6)$$

where $\beta = v_0/v_F$. From a simple comparison of eqs. (1) and (6) (and the help of eq. (4)), we obtain the final energy spectra of the graphene sheet under consideration (*i.e.*, on the xy plane under $\vec{H} = (0, 0, H)$ and $\vec{E} = (E, 0, 0)$):

$$\epsilon_{j,p_y} = \text{sgn}(j)(1 - \beta^2)^{3/4} \frac{\hbar v_F}{l_H} \sqrt{2|j|} + v_0 p_y, \quad (7)$$

where $l_H = \sqrt{\hbar c/eH}$ is the characteristic magnetic length. The expression above completely coincides with the expression obtained by solving the Dirac equation [26,27].

For the sake of comparison, a bilayer graphene (BG) is also considered. To this purpose, let us follow the same procedure as before. Thus, to go further, we must recall the energy spectrum of a BG in the vicinity of the Dirac point (analogously to eq. (5)) [4]:

$$\epsilon = \nu_b \left[\sqrt{\left(v_F^2 p^2 + \frac{t_{\perp}^2}{4} \right)} - \frac{\nu_{sb} t_{\perp}}{2} \right], \quad (8)$$

where t_{\perp} is the effective hopping energy between two layers (analogously to our recent contribution [9], we considered t_{\perp} close to 0.4 eV), and $\nu_{sb} = \pm 1$ is the bilayer sub-band index. As before, considering again a constant value of ϵ' , the quantized area on the momentum space reads now as

$$A(\epsilon') = \pi \frac{\epsilon'^2 + \epsilon' t_{\perp} + \beta^2 t_{\perp}^2 / 4}{v_F^2 (1 - \beta^2)^{3/2}}. \quad (9)$$

Again, a simple comparison of eqs. (1) and (9) (and the help of eq. (4)) leads us to the final energy spectra of the BG under consideration (*i.e.*, on the xy plane under $\vec{H} = (0, 0, H)$ and $\vec{E} = (E, 0, 0)$):

$$\epsilon_{j,p_y} = \text{sgn}(j) \sqrt{(1 - \beta^2)^{3/2} \frac{\hbar^2 v_F^2}{l_H^2} 2|j| + \frac{t_{\perp}^2}{4} (1 - \beta^2) + v_0 p_y}. \quad (10)$$

From those energy spectra, we are now able to focus on our aim: *quantum capacitance*, defined as [28–30]

$$C = -e^2 \int_0^{\infty} \frac{\partial f}{\partial \epsilon} \rho(\epsilon) d\epsilon, \quad (11)$$

where

$$f(\epsilon) = \frac{1}{\exp[(\epsilon - \mu)/k_B T] + 1} \quad (12)$$

is the Fermi-Dirac distribution function, μ is the chemical potential and

$$\rho(\epsilon) = -\frac{1}{\pi} \text{sgn}(\epsilon - \mu) \text{Im}\{G(\epsilon)\} \quad (13)$$

the density of states. Above, $G(\epsilon)$ is the single-particle Green function

$$G(\epsilon) = \sum_{j,p_y} \frac{1}{\epsilon - \epsilon_{j,p_y} + i \text{sgn}(\epsilon - \mu) \Gamma}, \quad (14)$$

where Γ is the width of the LLs related to the scattering on impurities. Thus, a minor calculation leads to

$$\rho(\epsilon) = \frac{1}{\pi} \sum_{j,p_y} \frac{\Gamma}{(\epsilon - \epsilon_{j,p_y})^2 + \Gamma^2}. \quad (15)$$

To go further, we have used the Poisson summation formula and have obtained

$$\rho(\epsilon) = \rho_0(\epsilon) + \rho_{osc}(\epsilon), \quad (16)$$

where

$$\rho_0(\epsilon) = \frac{L_y}{\pi^2 \hbar} \int_0^{p_{y\max}} dp_y \int_0^\infty \frac{\Gamma}{(\epsilon - \epsilon_{x,p_y})^2 + \Gamma^2} dx \quad (17)$$

and

$$\rho_{osc}(\epsilon) = \frac{2L_y}{\pi^2 \hbar} \operatorname{Re} \left\{ \sum_{k=1}^\infty \int_0^{p_{y\max}} dp_y \int_0^\infty \frac{\Gamma e^{-i2\pi kx}}{(\epsilon - \epsilon_{x,p_y})^2 + \Gamma^2} dx \right\}, \quad (18)$$

where L_y is the graphene size along the y -axis and $p_{y\max}$ is determined from the condition of the degeneracy of the LLS [26]:

$$0 < x = \frac{c}{eH} p_y < L_x. \quad (19)$$

From the above, it is easy to see that $p_{y\max} = eHL_x/c$.

Some assumptions now shall be done: $\mu \gg k_B T$ and $\Gamma \rightarrow 0$. The first one makes the integrand be significantly different from zero only near the point $\epsilon = \mu$; and, therefore, only energies $\epsilon \approx \mu$ are important for the magnetic oscillations. From these assumptions and after some evaluations (with the help of eqs. (1) and (3)), we obtain

$$\rho_0(\epsilon) = \frac{cL_y}{2\pi^2 \hbar^2 eH} \int_0^{eHL_x/c} dp_y m(\epsilon - v_0 p_y) \Theta(\epsilon - v_0 p_y - \epsilon_0), \quad (20)$$

where $\epsilon_0 = \epsilon_{j=0}$. Considering also $\mu \gg eEL_x = L_x v_0 \hbar / l_H^2$, the zero-field contribution to the density of states reads then as

$$\rho_0(\epsilon) \approx \frac{L_x L_y}{\pi \hbar^2} m(\epsilon). \quad (21)$$

Let us now focus our attention on the oscillating term of the density of states (eq. (18)). Considering again eqs. (1) and (3), as well as moving the integration over the energy variable, we obtain

$$\rho_{osc}(\epsilon) = \frac{2L_y}{\pi \hbar^2} \frac{c}{eH} \operatorname{Re} \left\{ \sum_{k=1}^\infty \int_0^{p_{y\max}} dp_y \int_0^\infty \delta(\epsilon - \epsilon_{x,p_y}) \times \exp \left[-ik \frac{c}{\hbar eH} A(\epsilon_x) \right] m(\epsilon_x) d\epsilon_x \right\}. \quad (22)$$

As mentioned above, the relevant values are those that satisfy $\epsilon \approx \mu$; and, therefore, we expand the function $A(\epsilon)$ in the vicinity of μ :

$$A(\epsilon) \approx A(\mu) + 2\pi m(\mu)(\epsilon - \mu). \quad (23)$$

Leading the above equation into eq. (22) we obtain

$$\rho_{osc}(\epsilon) = \frac{2L_y}{\pi \hbar^2} \frac{c}{eH} \operatorname{Re} \left\{ \sum_{k=1}^\infty \int_0^{p_{y\max}} dp_y \exp \left\{ -ik \frac{c}{\hbar eH} \left[A(\mu) + 2\pi m(\mu)(\epsilon - \mu - v_0 p_y) \right] \right\} m(\epsilon - v_0 p_y) \right\}, \quad (24)$$

since $\epsilon_{x,p_y} = \epsilon_x + v_0 p_y$ (see eqs. (7) and (10)). Previously, we have considered $\mu \gg eEL_x = L_x v_0 \hbar / l_H^2$ and, therefore, to simplify the solution, let us place this assumption only to $m(\epsilon - v_0 p_y)$, that resumes as $m(\mu)$. Further simple steps lead the oscillating term of the density of states to

$$\rho_{osc}(\epsilon) = \frac{2L_y}{\pi^2 \hbar v_0} \operatorname{Re} \left\{ \sum_{k=1}^\infty \frac{1}{k} \sin \left[k \frac{c\pi m(\mu)}{\hbar eH} v_0 p_{y\max} \right] \times \exp \left\{ -ik \frac{c}{\hbar eH} \left[A(\mu) + 2\pi m(\mu) \left(\epsilon - \mu - \frac{v_0 p_{y\max}}{2} \right) \right] \right\} \right\}. \quad (25)$$

A few more simple steps allow us to rewrite the above equation as

$$\rho_{osc}(\epsilon) = \frac{2L_y}{\pi^2 \hbar v_0} \sum_{k=1}^\infty \frac{1}{k} \sin \left(k \frac{\pi L_x m(\mu) v_0}{\hbar} \right) \times \cos \left\{ k \frac{c}{\hbar eH} \left[A(\mu) + 2\pi m(\mu) \left(\epsilon - \mu - \frac{v_0 p_{y\max}}{2} \right) \right] \right\}. \quad (26)$$

Considering again $\mu \gg eEL_x = L_x v_0 \hbar / l_H^2$, the above equation can be written as

$$\rho_{osc}(\epsilon) = \frac{2L_x L_y m(\mu)}{\pi \hbar^2} \times \sum_{k=1}^\infty \cos \left\{ \frac{kc}{\hbar eH} \left[A(\mu) + 2\pi m(\mu)(\epsilon - \mu) \right] \right\}. \quad (27)$$

From eqs. (11), (21) and (27), the quantum capacitance could be obtained in a closed form:

$$C = \frac{e^2 L_x L_y m(\mu)}{\pi \hbar^2} \left\{ 1 + 4 \sum_{k=1}^\infty x_k \frac{\cos \left[\frac{kc}{\hbar eH} A(\mu) \right]}{\sinh x_k} \right\}, \quad (28)$$

where

$$x_k = k \frac{2\pi^2 m(\mu) c k_B T}{\hbar eH}. \quad (29)$$

From the above result, it is easy to see that quantum capacitance oscillates as a function of the reciprocal magnetic field $1/H$; and the period of these oscillations depends on $A(\mu)$, that, on its turn, depends whether we consider either a MG or a BG. In this sense, quantum capacitance is depicted in fig. 1, for MG (a) and BG (b). Note that the period of the oscillations decreases by increasing the electric field; as well as the amplitude that, for any value of the applied electric field, decreases by decreasing the applied magnetic field. This result is of easy understanding, since an electric field changes the electronic cyclotron frequency, as highlighted by Peres and Castro [27]. A similar feature was found on graphene, on the oscillating magneto- and electro-caloric effects [9,10,15,17], as well as on the oscillating magnetic specific heat [25]. BG graphene behaves similarly; however, it oscillates faster than the MG, mainly due to the

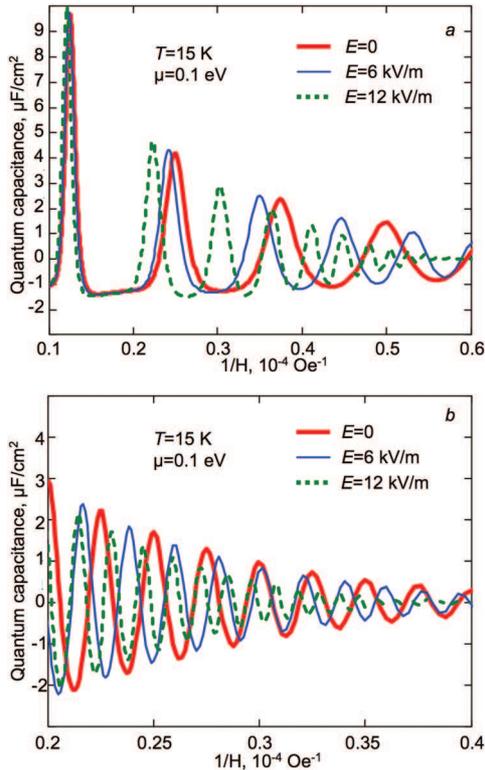


Fig. 1: (Colour online) Quantum capacitance for MG (a) and BG (b), both as a function of the reciprocal magnetic field $1/H$. Some values of the applied electrical field E are considered, as well as the temperature (15 K) and the chemical potential (0.1 eV, corresponding to 1200 Kk_B). The values of electric field E were chosen in such a way to satisfy $\beta < 1$.

hopping term, that also changes the electronic cyclotron frequency (see eq. (10)).

Considering that the argument of the cosine function in eq. (28) depends on $A(\mu)$, and this one, on its turn, depends also on the applied electrical field, it is natural to see oscillation as a function of E , as can be seen in fig. 2. Some values of the applied magnetic field are also presented and, in a similar fashion as previously discussed in fig. 1, this quantity (magnetic field) also rules the period of oscillations.

A remarkable point shall be addressed, as already discussed in ref. [15], and also found here for the quantum capacitance. If $\beta < 1$, we then have all of the above-described oscillations; however, if $\beta > 1$, the Landau structure collapses and the electronic motion becomes quasi-continuous. The consequence of this last case is the disappearance of the oscillations. In addition, the influence of the electric field on the Landau structure of graphene is much more remarkable than in other materials, like semi-conductors [31]. Thus, as a consequence, tuning of the magneto-electronic properties for device applications is indeed much more real for graphene.

Summarizing, quantum oscillations are a powerful tool to experimentally and theoretically study electronic properties of materials, namely energy spectra, scattering

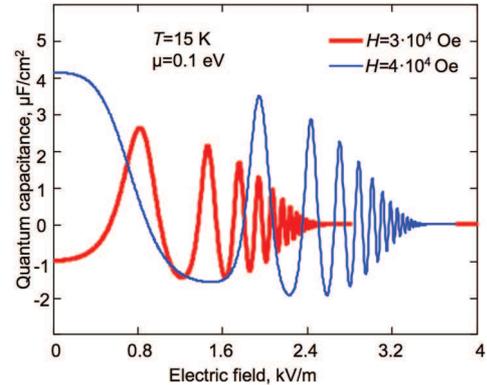


Fig. 2: (Colour online) Quantum capacitance for a MG as a function of the electric field. The aim of this figure is to stress the dependence of the quantum oscillations on the electric field. The parameters used are the same as those of fig. 1.

mechanisms, geometry of Fermi surface and much more information; and a deeper knowledge on these quantities gives to the scientific community a better understanding on some anomalous behaviors, such as Hall resistivity, thermopower, Nernst coefficient and many more. To crown the importance of this knowledge, these are the bases of several new applications of emerging functional materials, like graphene. In this direction, the present letter focuses on a thorough description of the quantum capacitance oscillation on monolayer and bilayer graphene under crossed electric and magnetic fields. We found a closed expression for quantum capacitance of MG and BG and, from these results, important parameters, like amplitude, period and phase, are highlighted as a function of electric and magnetic fields. We are convinced that these results are important to guide further experimental studies. Finally, our next step forward will be to consider the integer quantum Hall effect, taking into account the dependence of the Landau levels on the electric field, considering that this field is able to rebuild areas of localized states of electrons between these Landau levels.

ZZA declares that this work was supported by grants Nos. RFBR 15-02-03311a, PG MK-4471.2015.2, RSCF MKH-15-19-10049 and 3.1262.2014 from the Ministry of Education and Science of Russia. ZZA is also sincerely grateful to the Dmitry Zimin Foundation “Dynasty” for financial support. MSR acknowledges FAPERJ, CAPES, CNPq and PROPPI-UFF for financial support. The authors are deeply indebted to Prof. R. P. MEILANOV (in memoriam) due to his always helpful discussions on the topic discussed in this paper.

REFERENCES

- [1] KATSNELSON M. I., *Graphene: Carbon in Two Dimensions* (Cambridge University Press) 2012.

- [2] GOERBIG M. O., *Rev. Mod. Phys.*, **83** (2011) 1193.
- [3] FALCOVSKY L., *JETP*, **115** (2012) 1151.
- [4] FALCOVSKY L. A., *JETP Lett.*, **97** (2013) 429.
- [5] LUKŌYANCHUK I. A., VARLAMOV A. A. and KAVOKIN A. V., *Phys. Rev. Lett.*, **107** (2011) 016601.
- [6] NOVOSELOV K., GEIM A. K., MOROZOV S., JIANG D., KATSNELSON M., GRIGORIEVA I., DUBONOS S. and FIRSOV A., *Nature*, **438** (2005) 197.
- [7] GUSYNIN V. and SHARAPOV S., *Phys. Rev. Lett.*, **95** (2005) 146801.
- [8] LUKŌYANCHUK I. A., VARLAMOV A. A. and KAVOKIN A. V., *Phys. Rev. Lett.*, **107** (2011) 016601.
- [9] ALISULTANOV Z. and REIS M., *Solid State Commun.*, **206** (2015) 17.
- [10] ALISULTANOV Z., PAIXAO L. and REIS M., *Appl. Phys. Lett.*, **105** (2014) 232406.
- [11] PAIXÃO L., ALISULTANOV Z. and REIS M., *J. Magn. & Magn. Mater.*, **368** (2014) 374.
- [12] REIS M., *Phys. Lett. A*, **378** (2014) 1903.
- [13] REIS M., *Phys. Lett. A*, **378** (2014) 918.
- [14] REIS M., *J. Appl. Phys.*, **113** (2013) 243901.
- [15] REIS M. and SORIANO S., *Appl. Phys. Lett.*, **102** (2013) 112903.
- [16] REIS M., *Solid State Commun.*, **161** (2013) 19.
- [17] REIS M., *Solid State Commun.*, **152** (2012) 921.
- [18] REIS M., *Appl. Phys. Lett.*, **99** (2011) 052511.
- [19] REIS M., *Appl. Phys. Lett.*, **107** (2015) 102401.
- [20] ALISULTANOV Z. Z., *JETP Lett.*, **99** (2014) 232.
- [21] LIFSHITS I. and KOGANOV M., *Usp. Fiz. Nauk*, **69** (1959).
- [22] ALISULTANOV Z. Z., *JETP Lett.*, **99** (2014) 702.
- [23] LIFSHITZ I. and KOSEVICH A., *Sov. Phys. JETP*, **2** (1955) 636 (English Translation, 1956).
- [24] ONSAGER L., *London Edinburgh Dublin Philos. Mag. J. Sci.*, **43** (1952) 1006.
- [25] ALISULTANOV Z. and REIS M., arXiv preprint, arXiv:1508.06648 (2015).
- [26] LUKOSE V., SHANKAR R. and BASKARAN G., *Phys. Rev. Lett.*, **98** (2007) 116802.
- [27] PERES N. and CASTRO E. V., *J. Phys.: Condens. Matter*, **19** (2007) 406231.
- [28] GUSYNIN V., LOKTEV V., LUK'YANCHUK I., SHARAPOV S. and VARLAMOV A., *Low Temp. Phys.*, **40** (2014) 270.
- [29] ASGARI R., KATSNELSON M. I. and POLINI M., *Ann. Phys. (Berlin)*, **526** (2014) 359.
- [30] LOZOVIK Y. E., SOKOLIK A. and ZABOLOTSKIY A., *Phys. Rev. B*, **91** (2015) 075416.
- [31] ARONOV A. and PIKUS G., *Sov. Phys. JETP*, **22** (1966).