This item was submitted to Loughborough's Institutional Repository (<https://dspace.lboro.ac.uk/>) by the author and is made available under the following Creative Commons Licence conditions.

For the full text of this licence, please go to:
<http://creativecommons.org/licenses/by-nc-nd/2.5/>

Charging of graphene by magnetic field and mechanical effect of magnetic oscillations

Sergey Slizovskiy*

*Department of Physics, Loughborough University,
Loughborough LE11 3TU, UK and
Petersburg Nuclear Physics Institute, Russia*

We discuss the fact that quantum capacitance of graphene-based devices leads to variation of graphene charge density under changes of external magnetic field. The charge is conserved, but redistributes to substrate or other graphene sheet. We derive exact analytic expression for charge redistribution in the case of ideal graphene in strong magnetic field. When we account for impurities and temperature, the effect decreases and the formulae reduce to standard quantum capacitance expressions. The importance of quantum capacitance for potential Casimir force experiments is emphasized and the corresponding corrections are worked out.

PACS numbers: 75.70.Ak, 73.22.Pr, 12.20.Ds

Introduction. Graphene is a novel two-dimensional material having unique mechanical and electronic properties. The uniqueness of any two-dimensional material is that its electronic properties can be easily tuned by doping or gating. On top of that, graphene is the strongest known Quantum Hall material due to a sharp conical tip of its linear dispersion near the Dirac point. Magnetic oscillations can be noticed even at room temperature for magnetic field $B \gtrsim 5 \text{ T}^{1,2}$.

The electronic properties of materials show up in the Casimir effect. Namely, the effective action for electromagnetic field in the vacuum between materials depends on the boundary conditions that are determined by polarization tensor of the materials. In particular, at low distances the Casimir effect probes the spectrum of charged excitations up to rather high energies. There was a flurry of recent theoretical activity devoted to computation of Casimir effect for graphene³⁻⁷, with several controversies still unresolved: in particular, it is unclear what model to use for conductivity of graphene: Dirac model or hydrodynamic model, it may depend on the quality of the sample and on the distance between the bodies. Thus it is important to perform experiments to verify these computations. It is hard to make a mechanical measurement of Casimir force in graphene due to its two-dimensional nature and since it is almost always electrically charged. The electrostatic force is much stronger than the Casimir force, so, one needs to subtract the electrostatic contribution to single out the fluctuation-induced Casimir force. The first experiments have started to appear only recently⁸. Since graphene exhibits a strong Quantum Hall Effect (QHE) it would be of interest to repeat the experiments⁸ with strong transverse magnetic field.

A method for subtracting the (clearly dominant) electrostatic force⁹ was used in⁸: the electrostatic force depends on the gate voltage V as $F \sim (V - V_0)^2$, where V_0 is a residual graphene voltage due to

charged impurities and chemical potential difference with substrate. The formula has allowed the authors of⁸ to find the gate voltage V_0 where electrostatic force is fully compensated. The above formula does not include the quantum capacitance contribution, which is included by adding the “quantum capacitor” in a series connection: $\sigma = \int_{V_0}^V c_{total}(V') dV' = \int_{V_0}^V (c^{-1} + c_q^{-1}(V'))^{-1} dV'$ (σ is a charge density, $c = \epsilon/d$ is geometric capacitance per unit area and $c_q(V') = e^2 D(\mu_g(V'), T)$, where $D = dn/dE$ is a density of states^{2,10,11} is quantum capacitance). Then the electric pressure is $F/Area = \sigma^2/(2\epsilon)$, so we get

$$F/Area = \frac{1}{2\epsilon} \left(\int_{V_0}^V (c^{-1} + c_q^{-1}(V'))^{-1} dV' \right)^2 \quad (1)$$

$$\approx \frac{1}{2\epsilon} \left((V_0 - V)^2 c^2 - 2(V_0 - V) c^4 \int_{V_0}^V \frac{dV'}{e^2 D(\mu_g(V'), T)} \right)$$

where $D(\mu, T) = \int f'(E - \mu, T) D(E) dE$, $f(\mu, T)$ is a Fermi-Dirac distribution, $D(\mu)$ is a density of states for graphene. The chemical potential $\mu_g(V)$ for graphene depends on the applied voltage and the chemical doping may give a constant shift: $\mu_g(V) = -eV + const$. For ideal graphene $D(E) = \frac{2E}{\pi v_F^2}$ and so $D^{-1}(E, T)$ gives a singular contribution near the Dirac cone ($E = 0$) at small temperatures. Due to charge puddles in realistic graphene on substrate, the inverse density of states becomes smooth in the vicinity of the Dirac point², hence it gives a weakly V -dependent quantum capacitance of order 10^{-2} F/m^2 , thus the simple $(V - V_0)^2$ fit should work well for small intervals of $V - V_0$.

The story gets more interesting with magnetic field. The Casimir force for this case was estimated in Ref.7, where pronounced dependence on the magnetic field and the chemical potential was shown. Thus it makes sense to scan a wider range of chemical potentials in the experiment. The magnetic field does also influence the electrostatic force, since the charge of ideal graphene is a step-like function of chemical potential with size of the step depending

FIG. 1. Illustration of experimental setups

on the magnetic field value. Thus, even if we consider a suspended graphene with only chemical doping, its charge will oscillate when changing magnetic field. The discussion of electrostatic contribution in magnetic field and quantum capacitance effect is the aim of this note.

Below we consider three examples (see Fig.1):

- Graphene suspended over the wide trench etched in a metallic substrate, or, alternatively, it can be suspended by leaning on crests.
- Two sheets of graphene forming a capacitor with fixed voltage applied (such geometry was discussed in Ref.7 and argued to have a possibility of repulsive Casimir force)
- Graphene laying on the insulator-coated semiconductor with given gate voltage V_{gate} and with grounded parallel metallic plate (or sphere) hanging at the distance d over graphene (actually, it is attached to vibrating cantilever of atomic-force microscope). Such geometry was used in the recent experiment⁸.

Below we derive explicit analytic formulas for the case of ideal graphene at zero temperature and then discuss a more realistic situation with the approach similar to Ref.2 and 12. The effect we discuss below was essentially observed experimentally in the context of pinning of $\nu = 2$ filling factor in QHE experiments with strong quantum capacitance effect¹³ and led to significant increase in precision for Hall resistance measurement.

With magnetic field the energy levels of conduction band of graphene are $E_k = \text{sign}(k)\sqrt{\alpha|kB|}$ where $\alpha = 2\hbar v_F^2$ and k is an integer, with degeneracy $g_s g_v C|B|$ per unit area, where $C = \frac{e}{2\pi\hbar}$ and $g_v = 2$, $g_s = 2$ accounts for spin and valley degeneracy.

When the levels are quantized, only the levels below the chemical potential μ_g would be filled. For

undoped graphene one would have half-filled zero LL, this serves as a reference point for summation of formally infinite spectrum of “Dirac sea”. For generic μ the charge density is quantized and given by

$$n(B) = 4C|B| \left(\left\lfloor \frac{\mu_g^2 \text{sign}(\mu_g)}{\alpha|B|} \right\rfloor + 1/2 \right) \quad (2)$$

where $\lfloor \cdot \rfloor$ denotes the integer part (Floor).

Consider the case of graphene suspended over the etched trench of depth d in a metallic substrate. In this geometry graphene is connected to a conductor. Another example to which the same computation applies is a piece of pyrographite from which a large graphene flake has exfoliated.

Consider graphene having the chemical potential μ_{g0} for mobile carriers with density n_0 at zero magnetic field. These are related as

$$n_0 = \frac{1}{\pi} \text{sign}(\mu_{g0}) \left(\frac{\mu_c}{\hbar v_F} \right)^2 = 4 \text{sign}(\mu_{g0}) \frac{C}{\alpha} \mu_{g0}^2 \quad (3)$$

When the magnetic field is switched on, the electronic structure of graphene changes much stronger than the one for the other materials involved, so, we consider the effect of magnetic field only on graphene and thus the chemical potential of the conductor in the bottom of the trench is fixed (here we neglect the electric penetration depth for the conductor). Since the magnetic field may induce changes of the carrier number of graphene, $n = n_0 + \delta n$, this creates an extra electric field $\delta E = -e\delta n/\epsilon$ which shifts the chemical potential of graphene by $-e d \delta E$, so we solve

$$\mu_g - \mu_{g0} = e^2 \delta n / c \quad (4)$$

where δn depends on B and $c = \epsilon/d$ is a capacitance per unit area: d is the distance between the plates of the capacitor and ϵ is a dielectric permittivity ($\epsilon = \epsilon_0$ for the vacuum). Using Eq.(2) we get the equation:

$$4C e^4 \text{sign}(\mu_{g0}) \delta n^2 + (\alpha c^2 + 8C c e^2 |\mu_{g0}|) \delta n + 4C c^2 |B| \alpha \left(\left\{ \frac{\mu_{g0}^2 \text{sign}(\mu_{g0})}{\alpha|B|} \right\} - \frac{1}{2} \right) = 0, \quad (5)$$

which has a simple solution in the limit $\delta n \ll n_0$:

$$\delta n = \frac{4C|B|}{1 + 8C|\mu_{g0}|e^2/(\alpha c)} \left(\frac{1}{2} - \left\{ \frac{\mu_{g0}^2 \text{sign}(\mu_{g0})}{\alpha|B|} \right\} \right) \quad (6)$$

where $\{x\} = x - [x]$ gives the fractional part. The exact solution is also straightforward. This result shows how the charge of graphene oscillates when the magnetic field is changed, see the dotted curve in Fig.2. The corresponding force oscillation follows from $F/\text{Area} = e^2 n^2 / (2\epsilon)$.

It is clear that temperature and disorder would reduce the effect we discuss. For the case of very clean

FIG. 2. Change of electron density (m^{-2}) when turning on the magnetic field ($B = 6 \text{ T}$) as a function of initial chemical potential (first plot) and as a function of magnetic field (for $\mu_{\text{graphene}} = 50 \text{ meV}$), capacitance denominator is set here to 6. Dotted curves are for ideal graphene, solid curve accounts for Lorentzian broadening $5\sqrt{B(\text{T})} \text{ meV}$, dashed curve corresponds to broadening due to temperature $T = 300 \text{ K}$.

suspended graphene we expect the disorder to be weak and choose a simplified model of equal-shape broadening of all the Landau levels. It is clear that the actual result would depend mostly of the shape of the level that is nearest to μ_{g0} , so, it's the width of that level that we should take as our broadening. The broadening is computationally equivalent to smearing of the chemical potential, see Fig. 2.

The effect we discuss is another manifestation of integer Quantum Hall Effect. Qualitatively, if the last filled Landau level (LL) is less than half-filled, then the chemical potential is higher than the one without magnetic field, so, graphene wants to get rid of carriers and gets positively charged; the opposite happens for more-than-half filled level. This also shows that the upper bound for magnetic charging of graphene is half the population of one LL: $\frac{1}{2}4C|B|$, this bound is never achieved due to non-infinite geometric capacitance and level broadening. For example, with $d = 10 \text{ nm}$ and $\mu_{g0} = 20 \text{ meV}$ we get in the denominator: $1 + 8C|\mu_{g0}|e^2/(\alpha c) = 6.3$, which is a typical ratio of quantum to geometric capacitance for graphene experiments on thin insulator layers.

The magnetic oscillations of charge have a mechanical effect: the induced charge leads to attrac-

tion. Note that if graphene was initially charged at zero magnetic field (which typically does happen due to charged impurities), then the magnetic fluctuations could reduce the attraction. We see that a typical variation of electron density could be up to 10^{15} m^{-2} for $B \sim 6 \text{ T}$, which is comparable to charging due to impurities. This translates into the electric pressure

$$P = n^2 e^2 / (2\epsilon_0) \sim 2000 \text{ Pa} \quad (7)$$

The Casimir pressure at distance 10 nm between graphene and metal is estimated roughly as $f\hbar c\pi^2/(240d^4)$, where dimensionless f contains one power of QED fine-structure constant and equals roughly 0.026^3 . This gives a pressure of 3400 Pa . Note that the electrostatic force from the magnetic charging effect falls off as $1/d^3$ due to linearly decreasing capacitance and linearly decreasing capacitance, while the Casimir force falls off as $1/d^4$ at low temperatures (when $Td \ll \frac{\hbar c}{2\pi k_B}$), and as T/d^3 in the opposite limit. At room temperature, the low-temperature $1/d^4$ fall-off extends up to $d \sim 10^{-6} \text{ m}$, so, as the separation grows, the effect of magnetic fluctuations becomes dominant over the Casimir force. The electrostatic attraction due to impurities and difference in the work function decays as $1/d^2$, so it becomes dominant at larger separations.

Let us see if it is feasible to measure the effect. Consider a trench of width $l = 100 \text{ nm}$. Solving the elasticity equations one gets a parabolic form with tension $T = 1/2\sqrt{klP}$ and the central deflection is $h_0 = l^2 P/T/8 = l^{3/2} P^{1/2} k^{-1/2}$ where $k \approx 300 \text{ N/m}$ is a 2D Young modulus (note that it is possible that for small deformations the Hookes law is invalid for graphene due to microscopic out of plane buckled form of graphene^{14,15}, thus, for small deformations the effective Young modulus could be lower). For $l = 100 \text{ nm}$ trench we get $h_0 \approx 10^{-11} \text{ m}$, and it grows as $l^{3/2}$ if it is possible to make a wider trench. It is probably unrealistic to measure such tiny deflections due to dynamical rippling of suspended graphene. To remove the large effect of static rippling, one needs to measure the deflection of same sample at the same point while changing the magnetic field.

One could try to suspend graphene on elastic polymer, liquid crystal or liquid padding thus making it much easier for graphene membrane to deflect. The ideal situation would correspond to sliding-ends boundary conditions for graphene membrane and we need different equations involving bending rigidity to estimate the deflection: the central deflection is then given by $h_0 \approx \frac{5}{24} \frac{Pl^4}{\kappa}$ where $\kappa \approx 1 \text{ eV}$ is a bending rigidity of graphene. For $P = 2000 \text{ Pa}$ and $l = 100 \text{ nm}$ we get $h_0 = 260 \text{ nm}$, which is unreasonably large: graphene would just slide to the bottom of the trench. So, the truth must be between the two idealizations considered and moderate resistance to sliding may give the needed sensitivity.

Having the possibility to measure the electric at-

traction, one can apply voltage to graphene and tune it to minimize the attraction, analogously to Ref.8.

Now consider a geometry of *two parallel sheets of graphene*, that are electrically connected. This may be imagined as a drum made of two graphene sheets and can be experimentally realized by suspending two close-by graphene sheets over a deep trench.

Let these sheets be doped to μ_{10} and μ_{20} without magnetic field and have carrier densities n_{01}, n_{02} . An interest in such type of geometry stems from the prediction of possible repulsive Casimir force in magnetic field when μ_1 and μ_2 are of opposite signs⁷. We assume the carrier density redistribution in magnetic field: $n_{1,2}(B) = n_{0,1,2} \pm \delta n(B)$ and solve:

$$\mu_1 - \mu_{10} = \mu_2 - \mu_{20} + e^2 \delta n / c \quad (8)$$

together with Eq.(2) for both graphene sheets. The solution in the approximation $\delta n \ll n$ is

$$\delta n = 4C|B| \times \quad (9)$$

$$\times \frac{|\mu_{10}| \left(\left\{ \frac{\mu_{20}^2 \text{sign}(\mu_{20})}{\alpha|B|} \right\} - \frac{1}{2} \right) - |\mu_{20}| \left(\left\{ \frac{\mu_{10}^2 \text{sign}(\mu_{10})}{\alpha|B|} \right\} - \frac{1}{2} \right)}{|\mu_{10}| + |\mu_{20}| + 8C|\mu_{10}\mu_{20}|e^2/(\alpha c)}$$

where $c = \epsilon/d$. Note that the effect of magnetic oscillations cancels out if the two graphene sheets are at equal chemical potentials (and are of equal quality).

Now we turn to a much more flexible experimental setup used in⁸ and discuss *gated graphene laying on the insulator-coated semiconductor with a grounded parallel metal plate (or sphere) hanging over it*, the upper plate is an atomic force microscope used in frequency-shift regime^{8,16,17}. The presence of substrate and larger distance to the metal plate (of order 300 nm) makes the quantum capacitance effects weaker, but these are still important when compared to Casimir force. Remarkably, *this experimental setup allows for an excellent direct mechanical measurement of magnetic oscillations together with QHE*.

Due to larger distance between plates, we assume only weak magnetic oscillations, so the density of states is a smooth function and it is convenient to reformulate the solution in terms of continuum density of states: we have a series connection of two capacitors: the standard geometric one with $c_{geom} = \epsilon/d$ (per unit area) and a quantum one with $c_q = e^2 D(\mu_g)$, where $D = dn/dE$ is a density of states^{2,10,11}. So, the total capacitance is $c = (c_{geom}^{-1} + c_q^{-1})^{-1}$. We see that the relative effect of quantum capacitance decreases as $1/d$ due to decreasing of c_{geom} , so, for fixed voltage its contribution to the force decreases as $1/d^3$, which is small, but may still compete with the Casimir force that behaves as $1/d^4$ at low temperatures⁷.

To study magnetic oscillations and Casimir effect at strong magnetic field one needs to extend the experiment of Ref.8 by extra bottom-gating, so that a

FIG. 3. Density of states with Lorentz broadening 15 meV and carrier concentration dispersion $4 \cdot 10^{15} \text{ m}^{-2}$ as a function of chemical potential for $B = 5 \text{ T}$ (dotted), 10 T (dashed), 16 T (solid).

wide range of Landau level filling factors could be scanned (see fig. 1 c).

For the electrostatic force acting on the unit area of graphene we may use Eq.(1) and follow the model of Ref. 2 to get the density of states. The model consists of Lorentz and temperature level broadening superimposed on the Gaussian carrier number broadening due to charge puddles, see Fig. 3.

With the improved fit the value of residual potential difference V_0 can be mechanically measured with fabulous precision. V_0 is the potential difference between graphene and a metal plate when there is no electric field between them, so, it equals to graphene chemical potential:

$$V_0 = \mu_{\text{graphene}} + \text{const} \quad (10)$$

The electron doping of graphene is a linear function of bottom gate voltage (one can also easily write the quantum capacitance correction, but it is small for relatively thick insulator layer): $n \sim V_{\text{gate}}$. So, the experiment allows for precise measurement of both n and μ . Knowing this for the particular sample is also helpful for theoretical refinement of Casimir force computations. Importantly, the known $n(\mu, B)$ can be plugged back into Eq.(1) ($D = e^2 \frac{\partial n}{\partial \mu_{\text{graphene}}}$) to improve the fit and hence the precision. To get a more pronounced Quantum Hall physics, the above experiment may be repeated with gated graphene suspended over thin layer of insulator. Then one may hope to get a strong evidence for interaction effects.

To conclude, we have elaborated on the two possible experimental schemes to measure the Casimir effect for graphene with magnetic field. As a by-product, we note that the newly-developed mechanical method^{8,16,17} may lead to the precise measurement of density of states if sample is additionally gated.

Acknowledgements: I am grateful to Pablo Rodriguez-Lopez, Ignat Fialkovsky, Galina Klimchitskaya and Feo Kusmartsev for useful discussions.

This work has been supported by EPSRC through the grant EP/I02669X/1.

-
- * S.Slizovskiy@lboro.ac.uk
- ¹ A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, *Rev. Mod. Phys.* **81**, 109 (Jan 2009), <http://link.aps.org/doi/10.1103/RevModPhys.81.109>.
 - ² L. A. Ponomarenko, R. Yang, R. V. Gorbachev, P. Blake, A. S. Mayorov, K. S. Novoselov, M. I. Katsnelson, and A. K. Geim, *Phys. Rev. Lett.* **105**, 136801 (Sep 2010), <http://link.aps.org/doi/10.1103/PhysRevLett.105.136801>.
 - ³ M. Bordag, I. V. Fialkovsky, D. M. Gitman, and D. V. Vassilevich, *Phys. Rev. B* **80**, 245406 (Dec. 2009), [arXiv:0907.3242](http://arxiv.org/abs/0907.3242) [hep-th].
 - ⁴ I. V. Fialkovsky, V. N. Marachevsky, and D. V. Vassilevich, *Phys. Rev. B* **84**, 035446 (Jul. 2011), [arXiv:1102.1757](http://arxiv.org/abs/1102.1757) [hep-th].
 - ⁵ J. Sarabadani, A. Naji, R. Asgari, and R. Podgornik, *Phys. Rev. B* **84**, 155407 (Oct. 2011), [arXiv:1105.4241](http://arxiv.org/abs/1105.4241) [cond-mat.mes-hall].
 - ⁶ M. Bordag, G. L. Klimchitskaya, and V. M. Mostepanenko, *Phys. Rev. B* **86**, 165429 (Oct. 2012), [arXiv:1209.3302](http://arxiv.org/abs/1209.3302) [cond-mat.mtrl-sci].
 - ⁷ W.-K. Tse and A. H. MacDonald, *Physical Review Letters* **109**, 236806 (Dec. 2012), [arXiv:1208.3786](http://arxiv.org/abs/1208.3786) [cond-mat.mes-hall].
 - ⁸ A. A. Banishev, H. Wen, J. Xu, R. K. Kawakami, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, *Phys. Rev. B* **87**, 205433 (May 2013), <http://link.aps.org/doi/10.1103/PhysRevB.87.205433>.
 - ⁹ M. Bordag, G. L. Klimchitskaya, U. Mohideen, and V. M. Mostepanenko, *Advances in the Casimir Effect* (2009).
 - ¹⁰ S. Luryi, *Applied Physics Letters* **52**, 501 (Feb. 1988).
 - ¹¹ J. P. Eisenstein, L. N. Pfeiffer, and K. W. West, *Phys. Rev. B* **50**, 1760 (Jul 1994), <http://link.aps.org/doi/10.1103/PhysRevB.50.1760>.
 - ¹² S. Slizovskiy and J. J. Betouras, *Phys. Rev. B* **86**, 125440 (Sep. 2012), [arXiv:1203.5044](http://arxiv.org/abs/1203.5044) [cond-mat.mes-hall].
 - ¹³ T. J. B. M. Janssen, A. Tzalenchuk, R. Yakimova, S. Kubatkin, S. Lara-Avila, S. Kopylov, and V. I. Fal'ko, *Phys. Rev. B* **83**, 233402 (Jun 2011), <http://link.aps.org/doi/10.1103/PhysRevB.83.233402>.
 - ¹⁴ A. O'Hare, F. V. Kusmartsev, and K. I. Kugel, *Physica B Condensed Matter* **407**, 1964 (Jun. 2012).
 - ¹⁵ A. O'Hare, F. V. Kusmartsev, and K. I. Kugel, *Nano Letters* **12**, 1045 (Feb. 2012).
 - ¹⁶ C.-C. Chang, A. A. Banishev, R. Castillo-Garza, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, *Phys. Rev. B* **85**, 165443 (Apr 2012), <http://link.aps.org/doi/10.1103/PhysRevB.85.165443>.
 - ¹⁷ A. A. Banishev, C.-C. Chang, G. L. Klimchitskaya, V. M. Mostepanenko, and U. Mohideen, *Phys. Rev. B* **85**, 195422 (May 2012), <http://link.aps.org/doi/10.1103/PhysRevB.85.195422>.