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

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Received 29 July 2013, in final form 15 October 2013
Published 6 November 2013
Online at stacks.iop.org/JPhysCM/25/496007

Abstract

We discuss the fact that the 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 the substrate or other graphene sheets. We derive an exact analytic expression for charge redistribution in the case of ideal graphene in a strong magnetic field. When we account for impurities and temperature, the effect decreases and the formulas 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.

(Some figures may appear in colour only in the online journal)

1. Introduction

Graphene is a novel two-dimensional material which has 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 in its linear dispersion near the Dirac point. Magnetic oscillations can be noticed even at room temperature for magnetic fields $B \gtrsim 5\, T$ [1, 2].

The electronic properties of materials show up in the Casimir effect. Namely, the effective action of the electromagnetic field in the vacuum between materials depends on the boundary conditions which are determined by the polarization tensor of the materials. In particular, at low distances the Casimir effect probes the spectrum of charged excitations up to rather high energies; therefore, measurements of the Casimir effect contain information that is different from that obtained from ordinary conductivity measurements. There has been a flurry of recent theoretical activity devoted to computation of the Casimir effect for graphene [3–7], with several controversies still unresolved. In particular, it is unclear which model to use for the conductivity of graphene, the Dirac model or the hydrodynamic model; it may depend on the quality of the sample and 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 the 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 recently [8]. Since graphene exhibits a strong quantum Hall effect (QHE) it would be of interest to repeat the experiments [8] with a strong transverse magnetic field.

A method for subtracting the (clearly dominant) electrostatic force [9] was used in [8]. The electrostatic force depends on the gate voltage $V$ as $F \sim (V - V_0)^2$, where $V_0$ is the residual graphene voltage due to charged impurities and the chemical potential difference from the substrate. This formula has allowed the authors of [8] to find the gate voltage $V_0$ where the 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: $\sigma = \int_{V_0}^{V} c_{\text{total}}(V')dV' = \int_{V_0}^{V} (c^{-1} + c_q^{-1}(V'))^{-1}dV'\quad(\sigma$ is the charge density, $c = \epsilon/d$...
is the geometric capacitance per unit area and $c_g(V) = e^2D(\mu_g(V), T)$ is the quantum capacitance, with $D = d\rho/dE$ being the density of states [2, 10, 11]). Then the electric pressure is $F/\text{Area} = \sigma^2/(2\epsilon)$, so we get

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

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

where $D(\mu, T) = \int f'(E - \mu, T)D(E)dE$, $f(\mu, T)$ is the Fermi–Dirac distribution and $D(\mu)$ is the 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, $\mu_g(V) = -eV + \text{const}$. For ideal graphene $D(E) = \frac{2e^2}{\pi\hbar^2}$ 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 a substrate, the inverse density of states becomes smooth in the vicinity of the Dirac point [2]; 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 [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 also influences the electrostatic force, since the charge of ideal graphene is a step-like function of the chemical potential with the size of the step depending on the magnetic field value. Thus, even if we consider a suspended sheet of graphene with only chemical doping, its charge will oscillate when changing the magnetic field. A discussion of the electrostatic contribution in a magnetic field and the quantum capacitance effect is the aim of this paper.

Below we consider three examples (see figure 1).

- Graphene suspended over a 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 a geometry was discussed in [7] and it was argued that there was the possibility of a repulsive Casimir force).
- Graphene lying on an insulator-coated semiconductor with given gate voltage $V_{\text{gate}}$ and with a grounded parallel metallic plate (or sphere) hanging at the distance $d$ over the graphene (actually, it is attached to the vibrating cantilever of an atomic force microscope). Such a geometry was used in the recent experiment [8].

Below we derive explicit analytic formulas for the case of ideal graphene at zero temperature and then discuss a more realistic situation with an approach similar to [2, 12]. The effect we discuss below was essentially observed experimentally in the context of pinning of $v = 2$ filling factor in QHE experiments with a strong quantum capacitance effect [13] and led to a significant increase in precision for Hall resistance measurements.

2. Graphene–metal capacitor in a magnetic field

With a magnetic field the energy levels of the 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 $C = \frac{\pi e^2}{2m_0}$ and $g_s = 2, g_v = 2$ account for spin and valley degeneracy.

When the levels are quantized, only the levels below the chemical potential $\mu_g$ would be filled. For undoped graphene one would have a half-filled zero Landau level (LL); this serves as a reference point for summation of the formally infinite spectrum of the ‘Dirac sea’. For generic $\mu$ the charge density is quantized and is given by

$$n(B) = 4C|B| \left( \frac{\left| \frac{\mu g_0}{\beta C} \right| }{\alpha |B|} \right) + 1/2$$

where $[\ ]$ denotes the integer part (floor).

Consider the case of graphene suspended over an etched trench of depth $d$ in a metallic substrate. In this geometry the 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 been exfoliated.

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

$$n_0 = \frac{1}{\pi} \text{sign}(\mu_{g0}) \left( \frac{e^2}{\hbar v_F} \right)^2 = 4 \text{sign}(\mu_{g0}) C \frac{\mu_{g0}^2}{\alpha |B|}.$$  

When the magnetic field is switched on, the electronic structure of the graphene changes much more strongly than for the other materials involved; therefore, we consider the effect of the magnetic field only on the 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 the graphene by \(-e\delta E\), so we solve

\[
\mu_F - \mu_{g_0} = e^2 \delta n / c \tag{4}
\]

where \( \delta n \) depends on \( B \) and \( c = \epsilon / d \) is the capacitance per unit area; \( d \) is the distance between the plates of the capacitor and \( \epsilon \) is the dielectric permittivity (\( \epsilon = \epsilon_0 \) for vacuum). Using equation (2) we get the equation

\[
4C \epsilon^4 \text{sign}(\mu_{g_0}) \delta n^2 + (\alpha c^2 + 8C \epsilon e^2 |\mu_{g_0}|) \delta n
+ 4C \epsilon^2 |\mu_{g_0}| \left( \frac{\mu_{g_0} \text{sign}(\mu_{g_0})}{\alpha |B|} - \frac{1}{2} \right) = 0, \tag{5}
\]

which has a simple solution in the limit \( \delta n \ll n_0 \),

\[
\delta n = \frac{4C|B|}{1 + 8C|\mu_{g_0}| \epsilon^2 / (\alpha c)} \left( \frac{1}{2} - \frac{\mu_{g_0} \text{sign}(\mu_{g_0})}{\alpha |B|} \right) \tag{6}
\]

where \([x] = x - \lfloor x \rfloor\) 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 figure 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 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 on the shape of the level that is nearest to \( \mu_{g_0} \); therefore, it is the width of that level that we should take as our broadening. The broadening is computationally equivalent to smearing of the chemical potential, see figure 2.

The effect we discuss is another manifestation of the integer quantum Hall effect. Qualitatively, if the last filled LL is less than half-filled, then the chemical potential is higher than the one without magnetic field, so the graphene wants to get rid of carriers and becomes positively charged; the opposite happens for a more-than-half-filled level. This also shows that the upper bound for magnetic charging of graphene is half the population of one LL, \( \frac{1}{4} g_0 |B| \); this bound is never achieved due to non-infinite geometric capacitance and level broadening. For example, with \( d = 10 \) nm and \( \mu_{g_0} = 20 \) meV we get in the denominator \( 1 + 8C|\mu_{g_0}| \epsilon^2 / (\alpha c) = 6.3 \), which is a typical ratio of quantum to geometric capacitance for graphene experiments on thin insulator layers.

The magnetic oscillations of the charge have a mechanical effect: the induced charge leads to attraction. Note that if the graphene were initially charged at zero magnetic field (which typically happens 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 \( B \sim 6 \) T, which is comparable to the charging due to impurities. This translates into the electric pressure

\[
P = n^2 e^2 / (2\epsilon_0) \sim 2000 \text{ Pa}. \tag{7}
\]

The Casimir pressure at a distance of 10 nm between graphene and metal is estimated roughly as \( f h c \pi^2 / (240d^4) \), where dimensionless \( f \) contains one power of the 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 \( 1/d^2 \) due to linearly decreasing capacitance, while the Casimir force falls off as \( 1/d^3 \) at low temperatures (when \( Td \ll \hbar / k_B \)), and as \( T/d^3 \) in the opposite limit. At room temperature, the low-temperature \( 1/d^3 \) fall-off extends up to \( d \sim 10^{-6} \) 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 whether is feasible to measure the effect. Consider a trench of width \( l = 100 \) nm. Solving the elasticity equations one gets a parabolic form with tension \( T = 1/2 \sqrt{kP} \) and the central deflection is \( h_0 = FP/T = \beta^{1/2} pl^{1/2} k^{-1/2} \) where \( k \approx 300 \) N m\(^{-1}\) is the 2D Young modulus (note that it is possible that for small deformations Hooke’s law is invalid for graphene due to the microscopic out of plane buckled form of graphene [14, 15]; thus, for small deformations the effective Young modulus could be lower). For the \( l = 100 \) nm trench we get \( h_0 \approx 10^{-11} \) m, and it grows as \( \beta^{1/2} \) if it is possible to make a wider trench. It is probably unrealistic to measure such tiny deflections due to the dynamical rippling of suspended graphene. To remove the large effect of static rippling, one needs to measure...
the deflection of the same sample at the same point while changing the magnetic field.

One could try to suspend the graphene on an elastic polymer, liquid crystal or liquid padding, thus making it much easier for the graphene membrane to deflect. The ideal situation would correspond to sliding-end boundary conditions for the graphene membrane and we need different equations involving bending rigidity to estimate the deflection. The central deflection is then given by $h_0 \approx \frac{5Pd^4}{32\pi E}$ where $\kappa \approx 1$ eV is the bending rigidity of the graphene. For $P = 2000$ Pa and $l = 100$ nm we get $h_0 = 260$ nm, which is unreasonably large: the graphene would just slide to the bottom of the trench. Therefore, 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 attraction, one can apply voltage to the graphene and tune it to minimize the attraction, analogously to [8].

3. Graphene–graphene capacitor in a magnetic field

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 $\mu_{10}$ and $\mu_{20}$ without a magnetic field and have carrier densities $n_{01}, n_{02}$. The interest in this type of geometry stems from the prediction of a possible repulsive Casimir force in a magnetic field when $\mu_1$ and $\mu_2$ are of opposite signs [7] (note that the Casimir force between two graphene layers has two powers of the fine-structure constant, so it is significantly weaker). We assume the carrier density redistribution in the magnetic field $n_{1,2}(B) = n_{0,1,2} \pm \delta n(B)$ and solve

$$\mu_1 - \mu_{10} = \mu_2 - \mu_{20} + e^2 \delta n \mu$$

(8)

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

$$\delta n = 4C|B| \times \left( |\mu_{10}| \left( \frac{\mu_{20}^2 \text{sign}(\mu_{20})}{|\alpha|} - \frac{1}{2} \right) - |\mu_{20}| \left( \frac{\mu_{10}^2 \text{sign}(\mu_{10})}{|\alpha|} - \frac{1}{2} \right) \right) \left( |\mu_{10}| + |\mu_{20}| \right)^{-1} + 8C|\mu_{10}|\mu_{20}|^2/(\alpha c)^{-1}$$

(9)

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

4. Graphene–atomic force microscope capacitor in a magnetic field

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

Due to the 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 a continuum density of states. We have a series connection of two capacitors: the standard geometric one with $C_{\text{geom}} = \epsilon/d$ (per unit area) and a quantum one with $C_\text{q} = e^2/D_\mu$, where $D = d\mu/dE$ is the density of states [2, 10, 11]. Therefore, the total capacitance is $C = (C_{\text{geom}} + C_\text{q})^{-1}$. We see that the relative effect of the quantum capacitance decreases as $1/d$ due to decrease of $C_{\text{geom}}$, therefore, for fixed voltage its contribution to the force decreases as $1/d^2$, which is small, but may still compete with the Casimir force that behaves as $1/d^4$ at low temperatures [7].

To study the magnetic oscillations and the Casimir effect at strong magnetic fields one needs to extend the experiment of [8] by an extra bottom-gating, so that a wide range of Landau level filling factors can be scanned (see figure 1(c)).

For the electrostatic force acting on a unit area of graphene we may use equation (1) and follow the model of [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 figure 3.

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

$$V_0 = \mu_{\text{graphene}} + \text{const.}$$

(10)

The electron doping of the graphene is a linear function of the bottom gate voltage (one can also easily write the quantum
capacitance correction, but it is small for a relatively thick insulator layer, \( n \sim V_{\text{gate}} \). Therefore, the experiment allows for precise measurement of both \( n \) and \( \mu \). 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 equation (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 a thin layer of insulator. Then, one may hope to get strong evidence for interaction effects.

5. Conclusions

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

Acknowledgments

I am grateful to Pablo Rodriguez-Lopez, Ignat Fialkovsky, Galina Klimchitskaya and Feo Kusmartsev for useful discussions and to Ryan Walastyan for proofreading. This work has been supported by the EPSRC through the grant EP/I02669X/1.

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