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Room-temperature quantum Hall effect in graphene: the role of the two-dimensional nature of phonons

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Abstract. We consider two-dimensional nature of the electron-phonon coupling in graphene as a source for the room-temperature quantum Hall effect discovered in 2007. It is shown that magnetic field introduces strong cut-off for coupling with the two-dimensional acoustic phonons, *viz.* the processes with energy tranfer exceeding $\hbar s l_B^{-1}$ are exponentially suppressed, while for three-dimensional phonons the cut-off is set by a temperature T (here s is the sound velocity and $l_B \propto B^{-1/2}$ is the magnetic length). Consequently, at sufficiently high temperatures and magnetic fields only a small part ($\sim \hbar s l_B^{-1}/T$) of the electron states is involved in coupling with a given electron state in comparison with the case of three-dimensional phonons. Hence, the percolation threshold is postponed, and the quantum Hall effect survives up to $T = 300\,\mathrm{K}$.

1. Introduction

The integer quantum Hall effect is known to be irrefutable proof of atomistically two-dimensional nature of the graphene sheets [1], produced originally by the exfoliation technique, and now by the epitaxial methods suitable for massive production of graphene. Though it was speculated to be "half-integer", in fact the Hall conductivity of graphene σ_{xy} is quantized in the units of e^2/h, while really intriguing is consequence of the plateaus, described by the redoubled odd numbers, so that $\sigma_{xy} = 2(2n + 1)$. This particular sequence is readily understood from the Landau level spectrum of graphene,

$$E_n = \mathrm{sgn}(n)\sqrt{2|n|}\hbar v_0 l_B^{-1}, \tag{1}$$

taking into account fourthfold degeneracy due to valleys and spin, and symmetry of the ground Landau level $n = 0$ with respect to zero energy point $E = 0$ (here $v_0 \approx 10^8\,\mathrm{cm/s}$ is the graphene effective electron velocity and $l_B = \sqrt{\hbar c/eB}$ is the magnetic length). In fact, the "half-integer" rule of quantization is not really universal, since $\sigma_{xy} = 0, \pm 4$, and even $\sigma_{xy} = \pm 1$ plateaus have been observed experimentally [2] at sufficiently low temperatures, and the latter set of plateaus ($\sigma_{xy} = \pm 1$) raises questions to a theory of Ref. [3], employing the non-linear σ-model with artificially added topological term to explain the "half-integer" effect.

However, the most striking feature of the quantum Hall effect in graphene is, in our opinion, its immunity to elevated temperatures, as high as 300 K [4], while 20 K is the best of what the traditional two-dimensional systems can boast. Although this fact has been considered by many researches just as manifestation of the large gaps between the ground ($n = 0$) and the first

excited ($n = \pm 1$) Landau levels, described by Eq. (1), physics behind the room-temperature quantum Hall effect in graphene is indeed much more exciting. Using Eq. (1) together with the usual expression for the cyclotron energy suitable for convenient two-dimensional structures, $\hbar\omega_c = \hbar eB/m^*c$, it is easy to check that the distances between $n = 0$ and $n = 1$ Landau levels of graphene and $InAs$ quantum wells ($m^* = 0.023m_0$) differ less than by a factor of 1.5 at $B = 30\,\mathrm{T}$, while the limiting temperatures differ by more than one order of magnitude. This means it is not just $T/\hbar\omega_c$ parameter that restricts observation of the quantum Hall effect at $T > 20\,\mathrm{K}$ in the convenient two-dimensional structures, but there are relevant inelastic scatterings, which destroy coherence in the electron subsystem, namely electron-electron and electron-phonon interactions. Though it is not easy to describe influence of the interactions on the quantum Hall effect explicitly, they are proved experimentally to limit maximum temperatures for observation of the effect, and the transition widths between adjacent plateaus are shown to depend on temperature as $\Delta B \propto T^\kappa$ with $\kappa \approx 0.42$, reflecting ideas of the so-called scaling theory of the integer quantum Hall effect [5]. In other words, dephasing processes at strong magnetic fields are rather tricky (compared to the case of zero magnetic field [6]) since almost all the electron states are localized and require self-consistent treatment at essentially non-zero temperature T, introducing heat bath of either photons or phonons [7].

In this paper we focus on the electron-phonon interaction in graphene as being more relevant at not too low temperatures [6], considering the two-dimensional acoustic phonons of graphene. Some time ago, the two-dimensional nature of the graphene phonons was shown theoretically [8] to change appreciably the temperature dependence of resistivity in the Bloch-Grüneisen regime, from $\rho \sim T^5$ to $\rho \sim T^4$, and this prediction was confirmed experimentally by Evefov and Kim [9]. And though graphene physics is far from being settled down, for example reported magnitude of the deformation potential Ξ varies from 3 to 30 meV (see discussion in Ref. [10]), we believe that the two-dimensional intrinsic graphene phonons dominate over the three-dimensional substrate phonons. Our results show that reduced dimensionality of the phonons is really dramatic at a strong magnetic field, which introduces strong cut-off for energy of a phonon to be absorbed or radiated. Notably, a crucial thing here is not the total rate of transitions from a given electron state (*i.e.* the inverse lifetime), but number of the electron states coupled with this state by phonons, which is reduced by factor $\hbar s l_B^{-1}/T \sim 10^{-1}$ in comparison with the case of three-dimensional phonons. This feature explains robustness of the quantum Hall in graphene to the temperatures, which are an order of magnitude higher than allowed for traditional two-dimensional structures with the essentially three-dimensional phonons.

2. Theory

It is well known that lateral dynamics of the electrons set on the Landau levels of graphene does not differ cardinally from the case of convenient two-dimensional electron gas, except for the multi-component (chiral) structure of the wave function [11]. However, the room-temperature quantum Hall effect in graphene exhibits only two plateaus, both belonging to the ground Landau level $n = 0$, and this particular case is reduced to the scalar one [11]. Therefore, we describe graphene electrons at a strong magnetic field using the model of Ref. [12], so the one-particle wavefunctions can be presented in basis of the "Landau stripes" Ψ_{nk} with $n = 0$, $\Psi_\alpha(\mathbf{r}) = \sum_k C_{\alpha k}\Psi_{0k}(\mathbf{r})$. To calculate the electron lifetime due to electron-phonon coupling, we apply the Fermi golden rule, and it is convenient to represent the total rate W_α for a given electron state α as a sum of the rates due to spontaneous emission $W_\alpha^{(1)}$, stimulated emission $W_\alpha^{(2)}$, and absorption $W_\alpha^{(3)}$ of the acoustic phonons, and each of them can be written as

$$W_\alpha^{(i)} = \frac{2\pi}{\hbar}\sum_{\beta\mathbf{Q}}(1 - f_\beta)|V_{\beta\alpha}|^2[n_Q]^{a_i}\delta(E_\alpha - E_\beta - b_i\hbar sQ), \qquad (2)$$

where $\beta \neq \alpha$ runs over all the electron states of the ground Landau level $n = 0$, $f_\beta = f_0(E_\beta)$ and $n_Q = n_0(\hbar sQ)$ are the Fermi-Dirac and Bose-Einstein distributions of the electrons and phonons, respectively, $a_1 = 0$, $a_2 = a_3 = 1$, $b_1 = b_2 = 1$, $b_3 = -1$, and $V_{\beta\alpha}$ represents matrix element of the electron-phonon interaction,

$$\hat{V} = \sum_{\alpha\beta\mathbf{Q}} i\Xi \sqrt{\frac{\hbar Q}{2s\rho_d \mathcal{V}_d}} P^{\mathbf{Q}}_{\beta\alpha} \hat{a}^+_\beta \hat{a}_\alpha, \tag{3}$$

$$P^{\mathbf{Q}}_{\beta\alpha} = \int d^3r \Psi^*_\beta(\mathbf{r}) e^{i\mathbf{Q}\mathbf{r}} \Psi_\alpha(\mathbf{r}). \tag{4}$$

Here Ξ is the deformation potential, s is the sound velocity, ρ_d and $\mathcal{V}_d$ are graphene/graphite density and normalization area/volume, depending on the dimensionality d (either 2 or 3). Essential difference between the cases of two-dimensional and three-dimensional phonons is hidden inside the sum in Eq. (2), which turns into the integral over either two-dimensional or three-dimensional phonon momentum $\mathbf{Q}$. To estimate the electron lifetime, we employ a model of uncorrelated random C-coefficients, which actually neglects the effects of localization, but has been found suitable for approximate treatment of the density of states and even σ_{xx} peaks [12]. Of course, this approximation makes $W^{(i)}_\alpha$ independent of the particular form of the eigenstate α, so only the energy dependence remains, and it is convenient to introduce the averaged rates

$$W^{(i)}(\varepsilon) = \overline{W^{(i)}_\alpha} = \frac{\sum_\alpha W^{(i)}_\alpha \delta(\varepsilon - E_\alpha)}{\sum_\alpha \delta(\varepsilon - E_\alpha)}, \tag{5}$$

where summation over all the electron states of the infinite system is implied, or, equivalently, disorder averaging is carried out. Under this approach only the averaged value of $|P^{\mathbf{Q}}_{\alpha\beta}|^2$ matters, and it is readily calculated using the relationships [12]

$$\int d^3r \Psi^*_{0k_1}(\mathbf{r}) e^{i\mathbf{Q}\mathbf{r}} \Psi_{0k_2}(\mathbf{r}) = \delta_{q_x, k_1 - k_2} e^{i\frac{k_1+k_2}{2} q_y l_B^2} e^{-\frac{q^2 l_B^2}{4}}, \tag{6}$$

$$\sum_{k_1,k_2} \overline{C^*_{\alpha,k_1-k} C_{\alpha,k_2-k} C_{\beta,k_1} C^*_{\beta,k_2}} e^{i\gamma(k_1-k_2)} = N_L^{-1} \tag{7}$$

to be

$$\overline{|P^{\mathbf{Q}}_{\alpha\beta}|^2} = N_L^{-1} e^{-\frac{q^2 l_B^2}{2}}, \tag{8}$$

where $N_L = S/2\pi l_B^2$ is degeneracy of the Landau levels on the area S, and $\mathbf{q}$ is the in-plane component of the momentum $\mathbf{Q}$ (being generally three-dimensional). Eq. (8) shows that phonons with the in-plane momentum q exceeding l_B^{-1} couple electron states of the ground Landau level $n = 0$ exponentially weak. Thus, in the approximation neglecting possible corrections to W due to carrier localization, which are believed to be small, $W^{(i)}(\varepsilon)$ can be represented as

$$W^{(i)}(\varepsilon) = \frac{\pi\Xi^2}{\hbar s^2 \rho_d} \int \frac{d^dQ}{(2\pi)^d} [1 - f_0(\varepsilon - b_i\hbar sQ)] \mathcal{D}(\varepsilon - b_i\hbar sQ) [n_0(\hbar sQ)]^{a_i} \hbar sQ \exp\left(-\frac{q^2 l_B^2}{2}\right), \tag{9}$$

where $\mathcal{D}(\varepsilon)$ is the density of electron states, given for the ground Landau level $n = 0$ in case of the short-range disorder by exact solution due to Wegner [13]. Let us focus further on the regime of relatively high temperatures and magnetic fields, so that $\hbar s l_B^{-1}/T \ll 1$, assuming $T \ll \sqrt{2}\hbar v_0 l_B^{-1}, \Gamma$ at the same time (here Γ is the Landau level width). Typical value of $\hbar s l_B^{-1}/T$ at $T = 300\,\mathrm{K}$ and really strong magnetic field $B = 30\,\mathrm{T}$, required for the effect being observed

at this temperature, is 0.1 (using $s = 2 \cdot 10^6$ cm/s), so this case is quite relevant experimentally. Since we are really interested in electron states lying near the point of transition, with the energies $\varepsilon \sim T^\kappa$, the density of states in Eq. (9) can be taken outside the integral, reducing with good precision to the constant $\mathcal{D}_0 = \mathcal{D}(E = 0) \approx 0.36\Gamma^{-1}$ [13].

For two-dimensional phonons, the lifetime can be calculated under given conditions explicitly. The last factor in Eq. (9) introduces rather sharp cut-off of the integral at the inverse magnetic length l_B^{-1}, therefore the function $f_0(E)$, slowly varying on this scale, can be taken exactly at $Q = 0$, and T/E should be substituted for $n_0(E)$. The rest is easily calculated, yielding

$$W_{d=2}^{(i)} = (1 - f_0(\varepsilon))\frac{\Xi^2 \mathcal{D}_0 T}{2\rho_2 \hbar s^2 l_B^2}\left(\frac{\sqrt{\pi}\hbar s l_B^{-1}}{\sqrt{2}T}\right)^{1-a_i}, \tag{10}$$

so stimulated emission and absorption of phonons give equal contributions to the total rate, while a contribution of spontaneous emission is negligible. Such allotment is direct result of the magnetic length cut-off, since only "cold" phonons, with $\hbar s Q \ll T$ and, consequently, $n_0(\hbar s Q) \gg 1$, contribute to the process in the two-dimensional case.

The case of three-dimensional phonons is more complicated, since there is no magnetic length cut-off of the full phonon momentum Q, but only suppression at large Q, which competes with the three-dimensional phonon density of states. Technically, to evaluate the rates it is convenient to go to spherical coordinates, so that $q = Q \sin\vartheta$, and to integrate over the angles φ and ϑ first. The integral over ϑ reduces to the Dawson function $\mathcal{F}(x)$ with the well-known asymptotics,

$$\frac{1}{2}\int_0^\pi \exp\left(-\frac{t^2 \sin^2\vartheta}{2}\right)\sin\vartheta d\vartheta = \mathcal{G}(t) = \frac{\sqrt{2}\mathcal{F}(t/\sqrt{2})}{t} \sim \begin{cases} 1 - \frac{1}{3}t^2, & t \ll 1 \\ t^{-2}, & t \gg 1. \end{cases} \tag{11}$$

Although $\mathcal{G}(t)$ decays quadratically at large $t = Q l_B \gg 1$, this drop is completely compensated by three-dimensional density of the phonon states, therefore cut-off in Q is set by a temperature. This implies that the contributions of small $Q \ll l_B^{-1}$ can be neglected, and t^{-2} can be substituted for $\mathcal{G}(t)$ in further calculations. Hence, the only dimensionless parameter remains, namely $\delta = (\varepsilon - \mu)/T$, and all the contributions can be expressed as

$$W_{d=3}^{(i)} = C_i(\delta)\frac{\Xi^2 \mathcal{D}_0 T^2}{2\pi\rho_3 \hbar^2 s^3 l_B^2}, \tag{12}$$

where the coefficients C_i are integrated combinations of f_0 and n_0. For our purposes δ dependencies are not very important, and we choose the most natural case of $\delta = 0$, where the coefficients can be evaluated analytically, yielding $C_1 = \frac{\pi^2}{12}$, $C_2 = \frac{\pi^2}{24}$, $C_3 = \frac{\pi^2}{8}$. Finally, we obtain the ratio between the full rates W at $\varepsilon = \mu$ for two different phonon dimensionalities,

$$w = \frac{W_{d=3}}{W_{d=2}} = \frac{\pi}{4}\frac{Td}{\hbar s}, \tag{13}$$

where $d = \rho_2/\rho_3 \approx 0.34\,\text{nm}$ is the interlayer distance of graphite. Though w grows with a temperature, at $T = 300\,\text{K}$ it is equal to 0.5, so the lifetime due to the two-dimensional phonons is half of that of the three-dimensional case. Hence, the electron-phonon interaction turns out to be even stronger in the two-dimensional case in the sense of the electron lifetimes. However, it more important how much electron states the interaction mixes with a given one than how intensive the overall mixing is (like in any other percolation problem), and this aspect, vital for the room-temperature quantum Hall effect, is discussed in the final section.

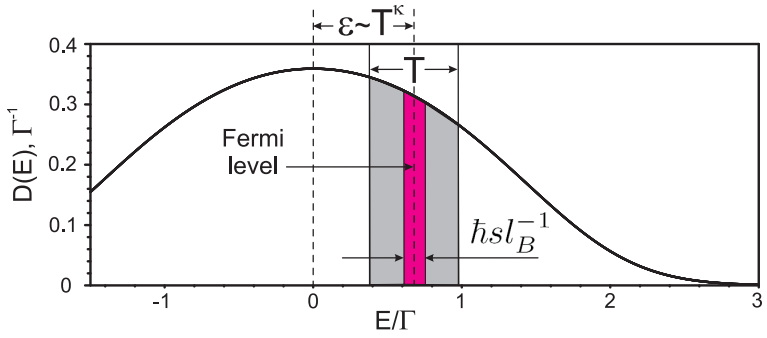

Figure 1. Electron states on the ground Landau level of graphene, producing fundamental plateaus of the integer quantum Hall effect ($\sigma_{xy} = \pm 2$). Active mixing of the electron states by the two-dimensional phonons takes place inside $\hbar s l_B^{-1}$ wide stripe, which is substantially narrower than the temperature stripe, germane to the case of three-dimensional phonons.

3. Discussion and conclusions

Let us recall qualitative picture of the quantum Hall transitions (occurring between the plateaus), using density of the electron states on the ground Landau level of graphene, as sketched in Fig. 1. In order to ascertain magnitude of the conductivity σ_{xx} at given position of the Fermi level μ, we have to consider a kind of percolation problem [7], where localized electron states (α, β, *etc.*) are treated as nodes of the resistor network, connected by the links (conductivities) with strengths, proportional to the rates $W_{\alpha\beta}$ due to electron-phonon interaction, which are given by Eq. (2) with omitted summation over the final states ("β"). Indeed, if we put electron into a certain state α, after the interval $\tau_\alpha = W_\alpha^{-1}$ it will be smeared over all the states β with non-exponentially small transition rates $W_{\alpha\beta}$, allowing transport within a cluster, formed from these states. Usually, number of the states involved in percolation is restricted by a temperature, so all of them lie inside the temperature stripe (shown by dim filling in Fig. 1), and the answer for percolation threshold in this case is well known, $\varepsilon \sim T^\kappa$ [5]. However, two-dimensional phonons do not allow mixing of the electron states outside narrower stripe of the width $\hbar s l_B^{-1}$ (dark area in Fig. 1), hence the threshold is postponed to $\varepsilon \sim (\hbar s l_B^{-1})^\kappa$. This means that the integer quantum Hall effect at high magnetic fields is not restricted by temperature at all, provided that phonons are strictly two-dimensional. Of course, three-dimensional substrate phonons must spoil this picture, but they seem to be suppressed by the two factors, namely weakness of the interlayer chemical links and strong magnetic field, as high as 45 T [4].

Acknowledgments

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