THz response of nonequilibrium electrons of highly doped graphene on a polar substrate

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Abstract. We consider high-frequency response of a system consisting of drifting electrons in a highly doped graphene and surface polar optical phonons of a polar substrate. We obtain the dielectric function, the frequencies and the decrement/increment of cooperative plasmon–optical phonon oscillations for this interacting system. We find that the response depends significantly on the degree of nonequilibrium for the electrons. In particular, the interaction between drifting plasmons and surface polar optical phonons leads to instability of the electron subsystem due to Vavilov–Cherenkov effect. We suggest that the hybrid system, a graphene on a polar substrate, is capable of using in amplifiers or generators of THz electromagnetic radiation.

Keywords: graphene, plasmon, optical phonon, instability, dielectric function

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1. Introduction

As is well known [1], graphene shows a number of remarkable physical properties. In particular, the kinetic energy E_k of free carriers as a function of momentum module $|\mathbf{p}| = p$ is linear in a wide energy range [2]:

$$E_k = \pm v_F p\,, \tag{1}$$

where $v_F \approx 10^8$ cm/s is the Fermi velocity (the signs '+' and '–' in Eq. (1) correspond to electrons and holes, respectively). This symmetry of energy bands allows considering the graphene as a bipolar semiconductor. To work only with electrons, one should have the concentration of electrons much greater than that of holes. This is can be achieved by doping the graphene. It can be doped chemically or by applying some positive electric voltage to a gate, which is generally placed under a substrate. In addition, one can use a so-called electrolytic gate [3] in order to achieve very high electron concentrations (up to 4×10^{14}cm^{-2}).

The influence of substrate on the properties of electrons in graphene has been studied in the works [4–10], including the case of nonequilibrium electrons. However, optical properties and cooperative plasma oscillations in highly doped graphene, as well as surface polar optical phonons (SPOP) arising from a polar substrate have been studied only for the equilibrium electrons [11–15]. In the present study, we calculate dielectric function of the system mentioned above for the case of nonequilibrium electrons. Basing on this dielectric function, we study oscillations of the coupled system. Although similar studies have been reported earlier [16, 17], they refer to materials where free electrons have a parabolic energy spectrum.

2. Basic equation and model criteria

At first, let us choose a coordinate system and consider an electronic subsystem. Let Oz axis be perpendicular to the plane of graphene sheet. Then axes Ox and Oy would belong to the plane of graphene. The graphene sheet is situated on a polar substrate at $z = 0$. The electronic subsystem can be described by a kinetic equation for distribution function $F(\mathbf{r},\mathbf{p},t)$ of electrons in highly doped graphene:

$$\frac{\partial F}{\partial t} + v_F \frac{\mathbf{p}}{p}\frac{\partial F}{\partial \mathbf{r}} - e\mathbf{E}\big|_{z=0}\frac{\partial F}{\partial \mathbf{p}} = I\{F\}\,. \tag{2}$$

Here $\mathbf{r} = (x, y)$ and $\mathbf{p} = (p_x, p_y)$ are respectively the coordinates and the momenta, $-e$ is the electron charge ($e > 0$), t the time, and $I\{F\}$ the collision integral. The total electric field $\mathbf{E}(\mathbf{r},z,t) = \mathbf{E}_0 + \mathbf{E}_s(\mathbf{r},z,t)$ consists of an external stationary homogeneous driving electric field $\mathbf{E}_0$ and a self-consistent field $\mathbf{E}_s(\mathbf{r},z,t)$ of electrons. The total field in Eq. (2) should be calculated in the plane of graphene, i.e. at $z = 0$.

In the case of highly doped graphene, the electron concentration is high and the main mechanism of collision corresponds to electron-electron collisions [5, 18, 19]. Then the stationary spatially uniform solution of Eq. (2) is a shifted Fermi–Dirac distribution function:

$$f_0(\mathbf{p}) = \left(\exp\left(\frac{v_F p - v_0 p_x - \mu}{k_B T}\right) + 1\right)^{-1}, \tag{3}$$

where the drift velocity $\mathbf{v}_0 = (v_0, 0)$ is directed along the Ox axis. Here k_B and T are respectively the Boltzmann constant and the electron temperature. The chemical potential μ can be determined from the normalisation condition

$$\frac{g_s g_v}{(2\pi\hbar)^2}\int d^2 p f_0 = n_0\,, \tag{4}$$

where $g_s = 2$ and $g_v = 2$ imply respectively the spin and valley degenerations, $\hbar$ is the reduced Planck constant, and n_0 the equilibrium surface concentration of electrons.

To describe the electron plasma oscillations, we will use a linear approximation $F(\mathbf{r},\mathbf{p},t) = f_0(\mathbf{p}) + f_1(\mathbf{r},\mathbf{p},t)$, where $f_1(\mathbf{r},\mathbf{p},t)$ represents a small perturbation to a function $f_0(\mathbf{p})$. We will neglect the collision integral in the linearised kinetic equation for $f_1(\mathbf{r},\mathbf{p},t)$:

$$\frac{\partial f_1}{\partial t} + v_F \frac{\mathbf{p}}{p}\frac{\partial f_1}{\partial \mathbf{r}} - e\mathbf{E}_0\frac{\partial f_1}{\partial \mathbf{p}} = e\mathbf{E}_s\big|_{z=0}\frac{\partial f_0}{\partial \mathbf{p}}. \tag{5}$$

For convenience we will use the scalar potential $\varphi(\mathbf{r},z,t)$ instead of the field $\mathbf{E}_s(\mathbf{r},z,t)$, which are linked by a simple relation $\mathbf{E}_s = -\nabla\varphi$.

Since Eq. (5) does not contain the coordinates and the time explicitly, the quantities $f_1(\mathbf{r},\mathbf{p},t)$ and $\varphi(\mathbf{r},z,t)$ can be represented as $f_1(\mathbf{r},\mathbf{p},t) = f_{\omega,\mathbf{k}}(\mathbf{p})e^{i\mathbf{kr}-i\omega t+i\cdot 0}$ and $\varphi(\mathbf{r},z,t) = \varphi_{\omega,\mathbf{k}}(z)e^{i\mathbf{kr}-i\omega t+i\cdot 0}$, where ω and $\mathbf{k}$ are the frequency and the wave vector of the perturbation, respectively. The infinitesimal term $i\cdot 0$ arises from the principle of causality. It gives the Landau rule to bypass the poles in the integrals (see below). The same rule applies if we add the infinitesimal collision integral f_1/τ at $\tau \to \infty$ to the r. h. s. of the kinetic equation (5).

the infinitesimal collision integral f_1/τ at $\tau \to \infty$ to the r. h. s. of the kinetic equation (5). Nevertheless, one should keep in mind that consideration of the case of a finite τ value does not make sense, since such a collision integral would violate the charge conservation law.

A neglect of the collision integral is valid if

$$\omega\tau_p >> 1, \tag{6}$$

where τ_p is the momentum relaxation time of electrons. Use of a classical kinetic equation for the electrons imposes the following restriction on the wave vector value:

$$k << k_F, \tag{7}$$

where $k_F = \sqrt{\pi n_0}$ is the absolute value of the equilibrium Fermi wave vector. If doping of the graphene is carried out by applying a positive voltage to the gate located under the substrate, our model would be valid if $kd >> 1$, where d is the substrate thickness, i.e. the distance between the gate and the graphene sheet.

We will also neglect the term $-e\mathbf{E}_0 \frac{\partial f_1}{\partial \mathbf{p}}$ in Eq. (5). This is valid if the changes in the electron momentum occurring in the field $\mathbf{E}_0$ over the time period of the wave are much smaller than the average electron momentum, and if the changes in the electron energy imposed by this field over the spatial period of the wave are much smaller than the average electron energy (i.e., the Fermi energy).

As follows from Eqs. (3) and (4), the chemical potential decreases with increasing drift velocity, so that we do not consider the drift velocity of electrons close to v_F. If the speeds typical for the electrons are much smaller than the speed of light, Eq. (5) should be solved together with the Poisson equation for the Fourier components of the potential $\varphi_{\omega,\mathbf{k}}(z)$ and the necessary boundary conditions,

$$\begin{cases} \dfrac{d^2\varphi_{\omega,\mathbf{k}}}{dz^2} - k^2\varphi_{\omega,\mathbf{k}} = 0 \\ \varphi_{\omega,\mathbf{k}}\,|_{z\to\pm\infty} \to 0 \\ \varphi_{\omega,\mathbf{k}}\,|_{z=-\delta} = \varphi_{\omega,\mathbf{k}}\,|_{z=\delta} \\ \dfrac{d\varphi_{\omega,\mathbf{k}}}{dz}|_{z=\delta} - \varepsilon(\omega)\dfrac{d\varphi_{\omega,\mathbf{k}}}{dz}|_{z=-\delta} = \dfrac{4\pi e g_s g_v}{(2\pi\hbar)^2}\int d^2p f_{\omega,\mathbf{k}}(\mathbf{p}) \end{cases}. \tag{8}$$

Here $\delta \to +0$ and the dielectric permittivity $\varepsilon(\omega)$ involved in the last boundary condition is a function of frequency (see, e.g., [20]):

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)\omega_{TO}^2}{\omega_{TO}^2 - \omega^2 - i\gamma_{TO}\omega}, \tag{9}$$

where ε_0 and ε_∞ are respectively the static and high-frequency dielectric constants of the substrate, ω_{TO} is the frequency of transverse optical phonons, and the parameter γ_{TO} describes the phonon damping ($\gamma_{TO} << \omega_{TO}$).

Eqs. (3)–(5) and Eqs. (8)–(9) compose a basic system for the problem under consideration.

3. Results and discussion

Using the above system of equations, one can obtain the dielectric function for the interacting plasmons and substrate phonons:

$$\varepsilon(\Omega,\mathbf{K}) = 1+\Omega_e \frac{\Omega^2 - 1 + i\Gamma_{TO}\Omega}{\Omega^2 - \Omega_0^2 + i\Gamma_{TO}\Omega} \int \frac{d^2P}{\left(\Omega - \dfrac{\mathbf{KP}}{P} + i\cdot 0\right)} \frac{\mathbf{K}}{K}\frac{\partial f_0}{\partial \mathbf{P}}, \tag{10}$$

where for convenience the following dimensionless variables are introduced: $\Omega = \dfrac{\omega}{\omega_{TO}}$, $K = \dfrac{kv_F}{\omega_{TO}} = \sqrt{K_x^2 + K_y^2}$, $\Omega_e = \dfrac{4\pi e^2 g_s g_v k_B T}{(2\pi\hbar)^2 v_F \omega_{TO}(1+\varepsilon_\infty)}$, $\Omega_0 = \dfrac{\omega_{SO}}{\omega_{TO}}$, $\Gamma_{TO} = \dfrac{\gamma_{TO}}{\omega_{TO}}$, and $V_0 = \dfrac{v_0}{v_F}$.

Here $\omega_{SO} = \sqrt{\dfrac{1+\varepsilon_0}{1+\varepsilon_\infty}}\omega_{TO}$ denotes the frequency of surface optical phonons [13]. Notice that the dielectric function given by Eq. (10) is anisotropic if the electron drift is present. This could lead to rotation of the light polarisation plane. Also, if we put $\omega_{SO} = \omega_{TO}$ or $\varepsilon_0 = \varepsilon_\infty$, we obtain the dielectric function for the pure plasma oscillations in highly doped graphene, because the width of the residual radiation band is zero in this case.

It is well-known that the Landau collisionless damping is absent in highly doped graphene plasma whenever $k << k_F$. This is because the pure plasmon branch does not get to the region where the imaginary part of the dielectric function is nonzero. As shown for the equilibrium case [12], the interaction of plasmons with the SPOP leads to splitting of the pure plasmon branch into two branches, Ω_+ and Ω_-. The damping decrement Γ_+ corresponding to Ω_+ is equal to $-\Gamma_{TO}/2$, since this branch passes through the region where the imaginary part of the integral appearing in Eq. (10) is zero. From the other hand, the branch Ω_- does pass through the region where the imaginary part of the integral in Eq. (10) is nonzero. In this region both the term Γ_{TO} and the imaginary part of the integral involved in Eq. (10) give contributions to the Γ_- parameter. Thus, the nature of damping of the cooperative plasma and the SPOP oscillations for the Ω_- branch is the same as the nature of the Landau collisionless damping. As will be shown, the similar situation also occurs for the nonequilibrium case. However, starting from some values of the drift velocity and the $\mathbf{K}$ parameter, the damping effect is replaced by growing oscillations that correspond to electrical instability of our system.

When performing numerical calculations, we have chosen TlCl as a substrate material. This material [21, 22] has a very wide residual radiation band, since $\varepsilon_0 \approx 37.2$ and $\varepsilon_\infty \approx 5$ at the helium temperature. Moreover, we have $\omega_{TO} \approx 1.15\times 10^{13}\ \mathrm{s}^{-1}$ and $\Gamma_{TO} \approx 0.066$ for TlCl. We take the electron concentration $n_0 = 10^{13}\ \mathrm{cm}^{-2}$ and the electron temperature $T = 200$ K for our estimations. For the parameters chosen as above, the dimensionless equilibrium Fermi wave vector is equal to $K_F = \dfrac{v_F k_F}{\omega_{TO}} \approx 48.7$. The chemical potentials for the cases of $V_0 = 0$ and $V_0 = 0.4$ are $\mu \approx 50$ meV and $\mu \approx 41.6$ meV, so that we obtain $\dfrac{\mu}{k_B T} = 2.9$ and $\dfrac{\mu}{k_B T} = 2.4$, respectively. For

simplicity, we put $K_y = 0$. Further on, we will present the results for the values of parameters that satisfy the inequalities (6) and (7), and the other criteria of our model.

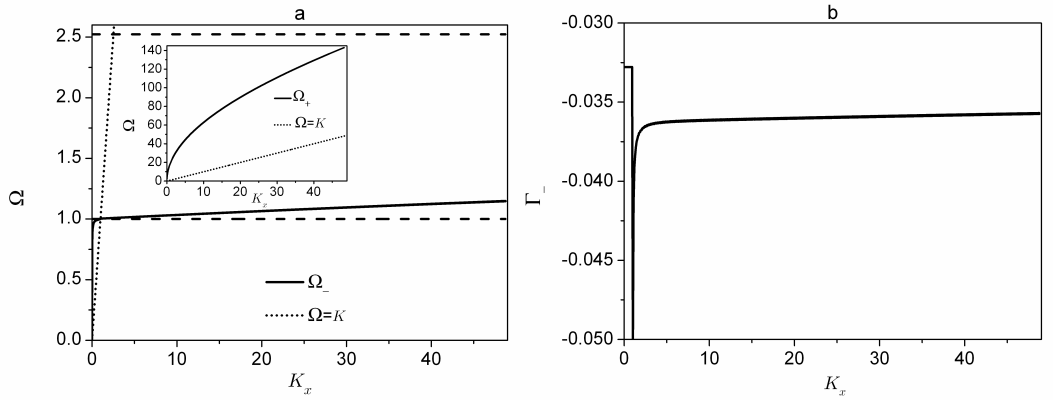

Fig. 1. (a) Frequency branch $\Omega_-(K_x)$ at $V_0 = 0$. Dash line restricts the residual radiation band. Branch $\Omega_+(K_x)$ is presented in the inset. (b) Decrement Γ_- as a function of K_x.

As seen from Fig. 1a, the frequency branch Ω_- passes close to $\Omega = 1$ and, due to a strong interaction, this branch never reaches the value $\Omega = \Omega_0 \approx 2.5$ corresponding to ω_{SO}, even at $K_x = K_F$. Since the branch Ω_- crosses the line $|\Omega| = K$, the damping caused only by the phonon damping corresponds to this branch in the region $\Omega > K$ (i.e., $\Gamma_- = -\Gamma_{TO}/2$). Therefore the total damping caused by the phonon damping and the Landau damping mechanisms corresponds to this branch in the region $|\Omega| < K$. A cutting behaviour of $\Gamma_-(K_x)$ can be explained by the fact that the imaginary part appearing in Eq. (10) has a point of discontinuity of the second kind at $|\Omega| = K$.

One should also notice a very rapid growth of the Ω_+ and Ω_- branches at small K_x. Such behaviour of the frequency branches leads to high group velocity of the wave in the region of the rapid growth. As is well known, the group velocity is related to energy transfer. Under electric field applied, a large part of the energy is given to almost dispersionless SPOP, of which group velocity is very small. As a result, the heat is not rejecting from the graphene. In the case of coupled vibrations of graphene plasmons and the SPOP, the group velocity of the waves corresponding to these vibrations is significantly greater. Hence, these waves could provide heat rejection from the graphene.

The both frequency branches for the nonequilibrium case (see Fig. 2) differ not very much from those shown in Fig. 1. From the other hand, the parameter Γ_- changes its sign, which corresponds to electrical instability of the system. In the absence of phonon damping, the Γ_- sign reverses strictly at $\Omega = V_0 K_x$, which corresponds to the Vavilov–Cherenkov effect. However, the total function Γ_- shifts by a half of the phonon damping parameter Γ_{TO} if Γ_{TO} remains nonzero (as in Fig. 2). It is worthwhile to notice that the effect of instability can be used for generating or amplifying the THz electromagnetic radiation.

When the electron drift is present, the electron temperature can be significantly higher than the ambient temperature. Our analysis shows that the effect of instability can also occur in the case of significant heating, whenever one can create high enough electron concentrations.

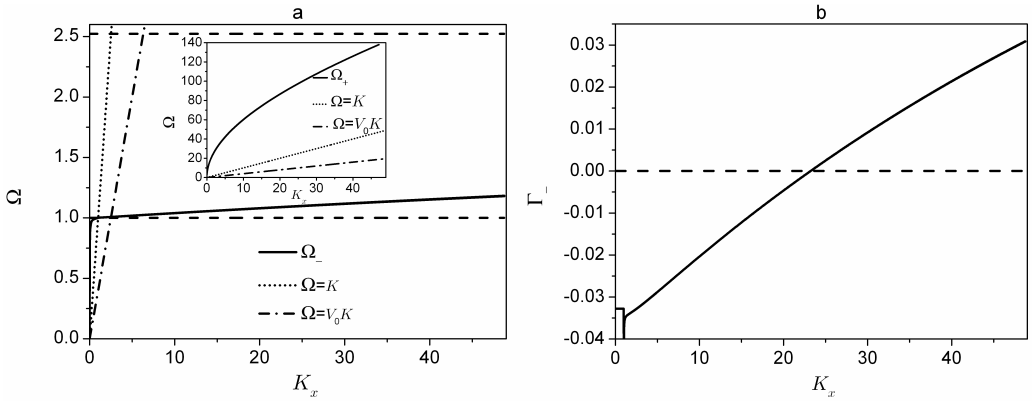

Fig. 2. The same dependences as shown in Fig. 1 calculated at $V_0 = 0.4$.

In conclusion, we have theoretically studied a hybrid system composed of a highly doped graphene and a polar substrate. We have found the THz response of the nonequilibrium electrons interacting with the SPOP. The response depends significantly on the electron drift velocity. In particular, the damping of oscillations of the system can give way to their growth, if the drift velocity is large. We suggest that the hybrid system under consideration is capable of using for amplification or generation of the THz electromagnetic radiation.

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Анотація. *Розглянуто надвисокочастотний відгук системи дрейфуючих електронів сильно легованого графену та поверхневих оптичних фононів полярної підкладки. Для взаємодіючої системи розраховано діелектричну функцію, частоти та декремент/інкремент спільних коливань. Показано, що відгук системи суттєво залежить від ступеня нерівноважності електронів. Зокрема, взаємодія між плазмонами та поверхневими оптичними фононами приводить до дрейфової нестійкості, зумовленої ефектом Вавілова–Черенкова. Розглянуту систему можна використовувати для генерації або підсилення терагерцового електромагнітного випромінювання.*